Ion-Irradiation of Tungsten: Modeling & Experiments

Presented by: SP Deshpande & P M Raole

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  - RCD BARC Mumbai: Mukherjee Saurabh, Pujari PK (Positron Spectroscopy)
  - FCD BARC Mumbai : Karki V, Singh Manish, Kannan S (SIMS)
  - RMD BARC Mumbai: Mishra S, Khan KB (High temp. high vac. annealing)
  - GGCU Bilaspur: Bajpai PK, Patel SP, Trivedi T (Boron beam)
  - UGC DAE Consortium Indore: Lakhani A (Low temp. resistivity)
  - IGCAR Kalpakkam: David C (ERDA)

Plan of Presentation

- Modeling (Presented by Shishir Deshpande)
    - Fusion Reactor Scenario: simplistic arguments
    - Defect Simulation
    - Cascade Analysis

- Experiments (Presented by PM Raole)
    - To be covered in the next presentation

Fusion Reactor Scenario

- For the divertor region, in a 500 MW ITER-like fusion reactor:
  - Average neutron flux on divertor ~$10^{12}/cm^2/s$; (peak 10^{13})
  - Number of PKA generated in tungsten ~ $10^{13}/cm^3/s$ (using ATTILA and SPECTER) (peak 10^{14})
- Taking 1 cm as a reasonable estimate of the tungsten-tile thickness implies a production of about ~$2.5x10^{13}$ Frenkel-Pairs/sec (using NRT) excluding in-cascade recombination
- MD simulations yield roughly 1/3rd of the above for an E_d ~90 eV (as FP recombination is taken care of), i.e., about $8x10^{12}$ FP/sec

Order of Magnitude Estimates

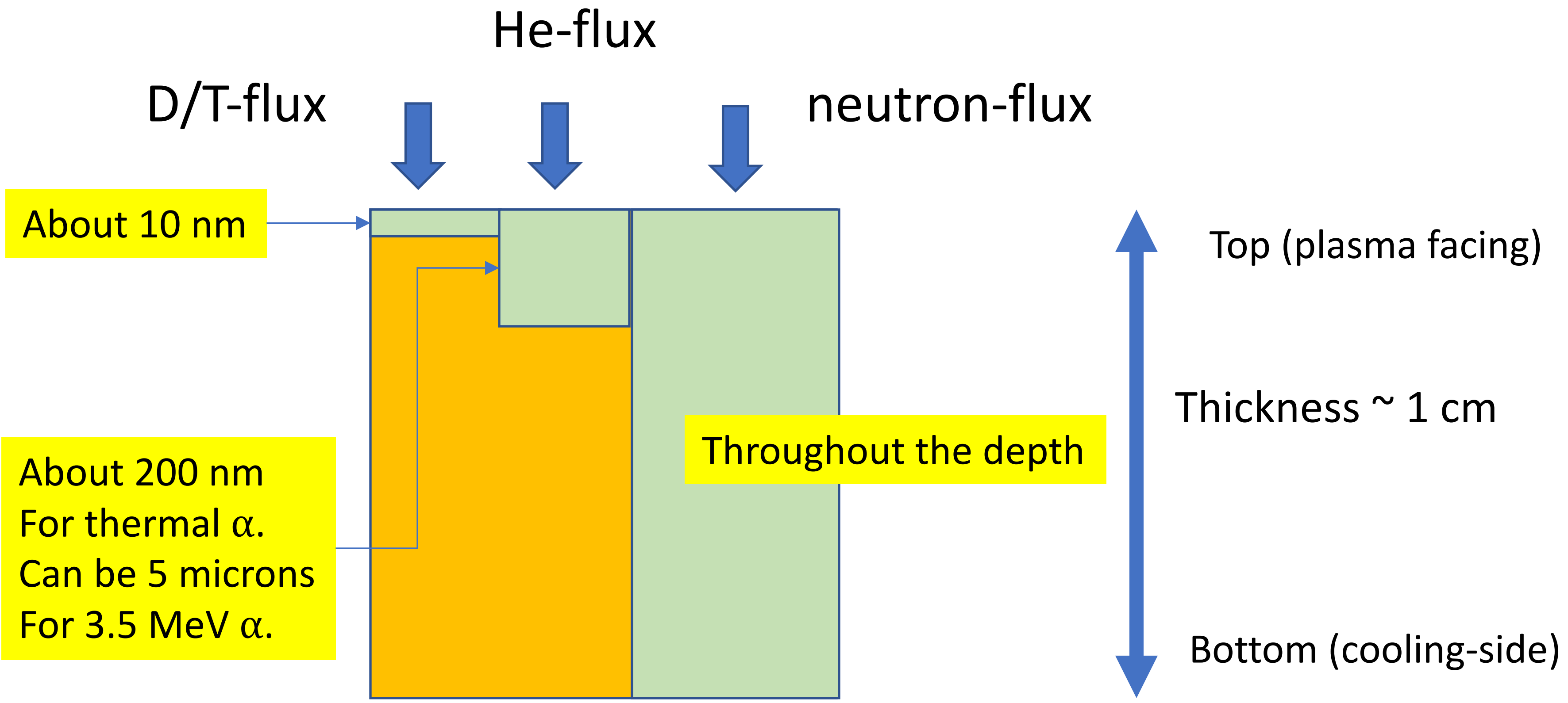
- Considering a 'wetted' area of 10 m^2 for a single-null divertor, with a thickness of 1 cm for tungsten, we get 8×10^{17} FP/s
- A point that needs to be mentioned here is that this will be uniform, throughout the thickness of the tungsten tile
- If all of the FP were to be considered as traps for a D or T atom and one considers a 'day-long' pulse (DEMO) then the inventory at the end of day would be ~ 7×10^{22} atoms or about 0.1 gm/day
- Of course these are very gross assumptions and one needs to carry out more detailed calculations of H-isotope retention in radiation-damaged tungsten

Concurrent Irradiation

- In a real power reactor, the fluxes due to D/T ions and He-ions will be concurrent with that of neutrons
- For 500 MW of fusion power, the power in α-particles will be about 100 MW. It corresponds to a generation rate of about 1.8×10^{20} α particles/s in the entire plasma volume. These particles are expected to slow down on the background plasma-ions and ultimately get exhausted
- In steady-state, to exhaust these He atoms through a narrow Scrape-off Layer channel surrounding the LCFS, the He-flux will be around (assuming 'wetted' area of 10 m^2) **$1.8 \times 10^{15}/cm^2/s$**
- Similarly, the D/T plasma flux can be estimated from divertor heat-flux (~10 MW/m^2) and typical edge temp. ~ 1keV, as **$3 \times 10^{18}/cm^2/s$**

With high D-flux, the top layer is going to saturate almost instantly. For D/T to be captured in deeper zones, it is necessary to have some diffusion mechanism and/or synergistic effects. He-particles can possibly contribute to this due to deeper spreading of the traps.

Study of radiation cascades has been carried out using MD to understand damage creation



Sketch (not to scale) of range of penetration

Penetration of Deuterium

- At the typical temperatures of edge plasma, the H-isotopes cannot directly penetrate deep into the bulk
- From the past work done by other CRP members, it is clear that the trapping of deuterium in tungsten happens due to the presence of defects (inherent + created by radiation damage)
- So it becomes important to find out how the D atoms reach the traps and whether they are able to access the traps in the deeper zone
- In one of our experiments we found that the D atoms had not penetrated enough into the substrate:
  - With a100 keV D-beam ($5x10^{17}/cm^2$) depth observed (350 nm) was less than that predicted by TRIM (600 nm). This possibly may be due to saturation of the top layer, preventing further ingress.

Molecular Dynamics Simulations

- We have considered a computational box containing 7 million tungsten atoms (single crystal)
- Code used: PARCAS [K. Nordlund PRL 77(4) 1996, p 699]
- A 64-core computational physics cluster at Institute for Plasma Research, Gandhinagar was used to simulate the dynamics up to 100-150 ps; it takes about 3-days to complete the run
- Potential selected was DNMD+ZBL2 (next slide, as elaborated in 2^{nd} RCM meeting)
- Self-atom (PKA) energies were chosen from 1-400 keV and each case was done with 100 bombardments

MD Simulations

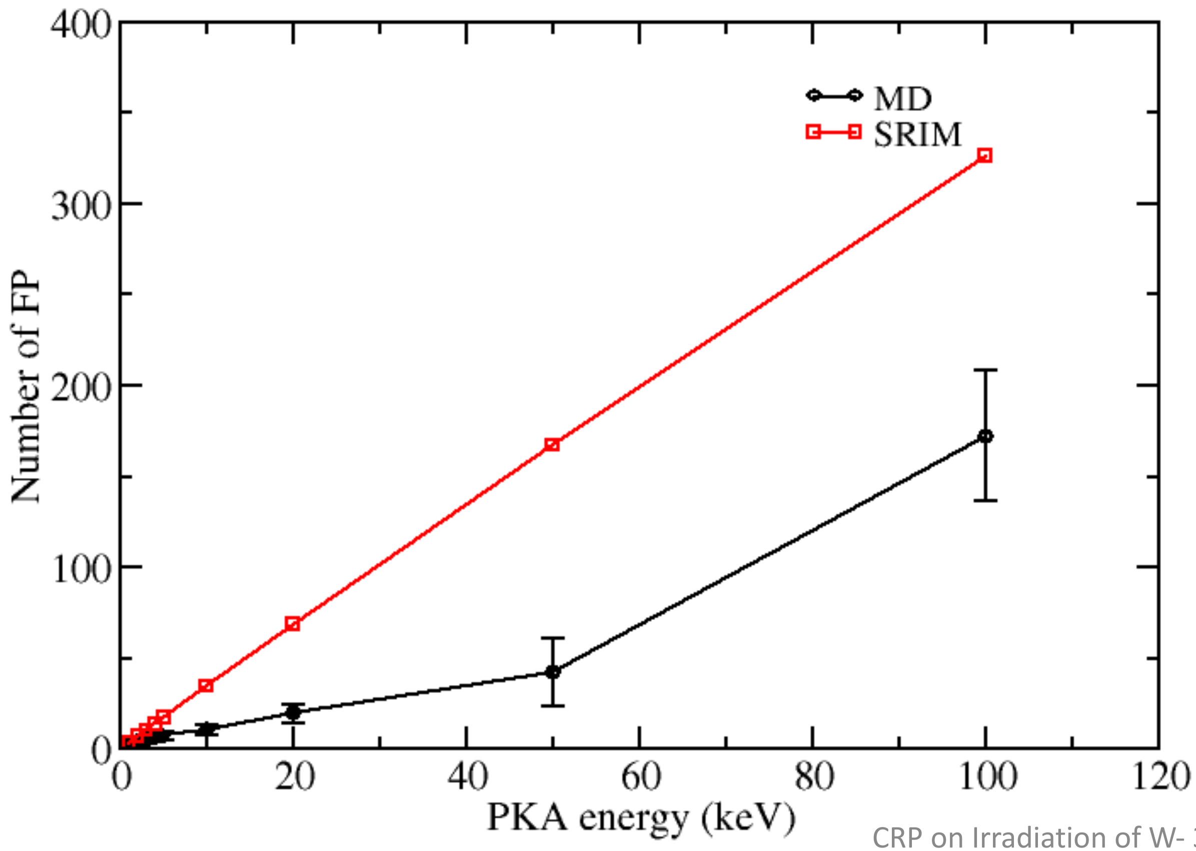
- Different potentials were compared earlier (2nd RCM)
- Selected DNMD+ZBL2 for further studies
- 1,2,3,4,5,10,20,30,50,100, 150, 200, 250, 400 keV PKA
- 9.8×10^5 atoms to 6×10^6
- 100 bombardments per energy (up to 100 keV so far)

	E_C (eV)	a (Å)	Elastic constants C_{ij} (Gpa)			E_V (eV)	E_I (eV)			
			C_{11}	C_{12}	C_{44}		E^d_{100}	E^d_{110}	E^d_{111}	E_{octa}
Expt.	8.90 [a]	3.1652 [b]	522.4 [b]	204.4 [b]	160.6 [b]	3.7			9.06	
DFT		3.18					11.513	9.84	9.55	11.7
EAM										
FS	-8.90	3.1652	522.44	204.41	160.61		8.652		7.805	8.524
AT	-8.90	3.1652	522.43	204.41	160.61		9.783		8.883	9.968
JW	-8.90	3.1652	522.47	204.45	160.64	3.554	10.277	10.157	9.50	10.393
DNMD	-8.90	3.1652	523.10	204.67	160.81	3.557	11.334	9.768	9.472	11.7
DNMD +ZBL1	-8.90	3.1652	523.10	204.67	160.81	3.557	11.332	9.766	9.472	
DNMD +ZBL2	-8.90	3.1652	523.10	204.67	160.81	3.557	11.333	9.768	9.472	
Zhou	-8.757	3.1652				3.567				
Tersoff										
Li	-8.86	3.1652	515	203	162	3.52	12.01	9.53	9.33	12.05
Juslin	-7.41	3.165	542	191	162	1.77	8.93	8.77	9.62	9.92

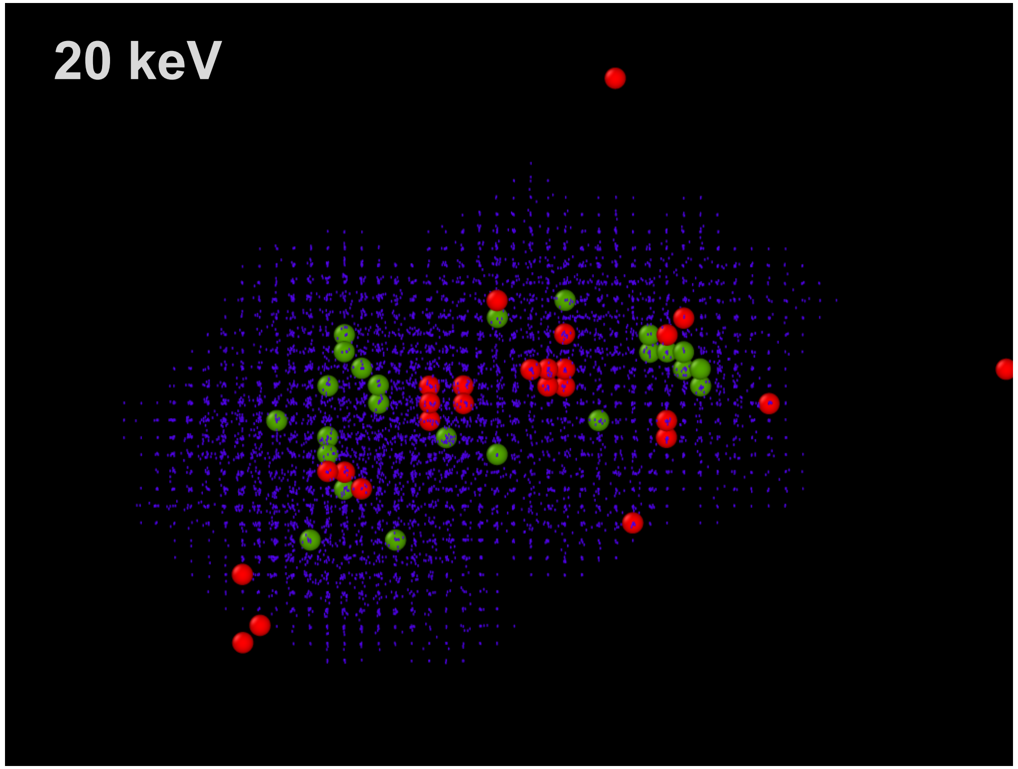
E_d (eV)	Expt [c]	AT	AT+ZBL	DNMD	DNMD+ZBL1	DNMD+ZBL2
Minimum	$\sim$ 40-50	64	48	55$\pm$ 3	55	41$\pm$ 1
Average	$\sim$ 80	166	128	88.3$\pm$0.7		84.5 $\pm$ 0.9

Clustering of interstitials and vacancies takes place while the cascade cools down. Shown at 20 ps after bombardment. At lower PKA energies, (E_{PKA} < 30 keV) smaller defect clusters are formed.

No. of FP generated as a function of PKA energy. Comparison with SRIM with Ed= 90 eV. ***Recombination is significant.***

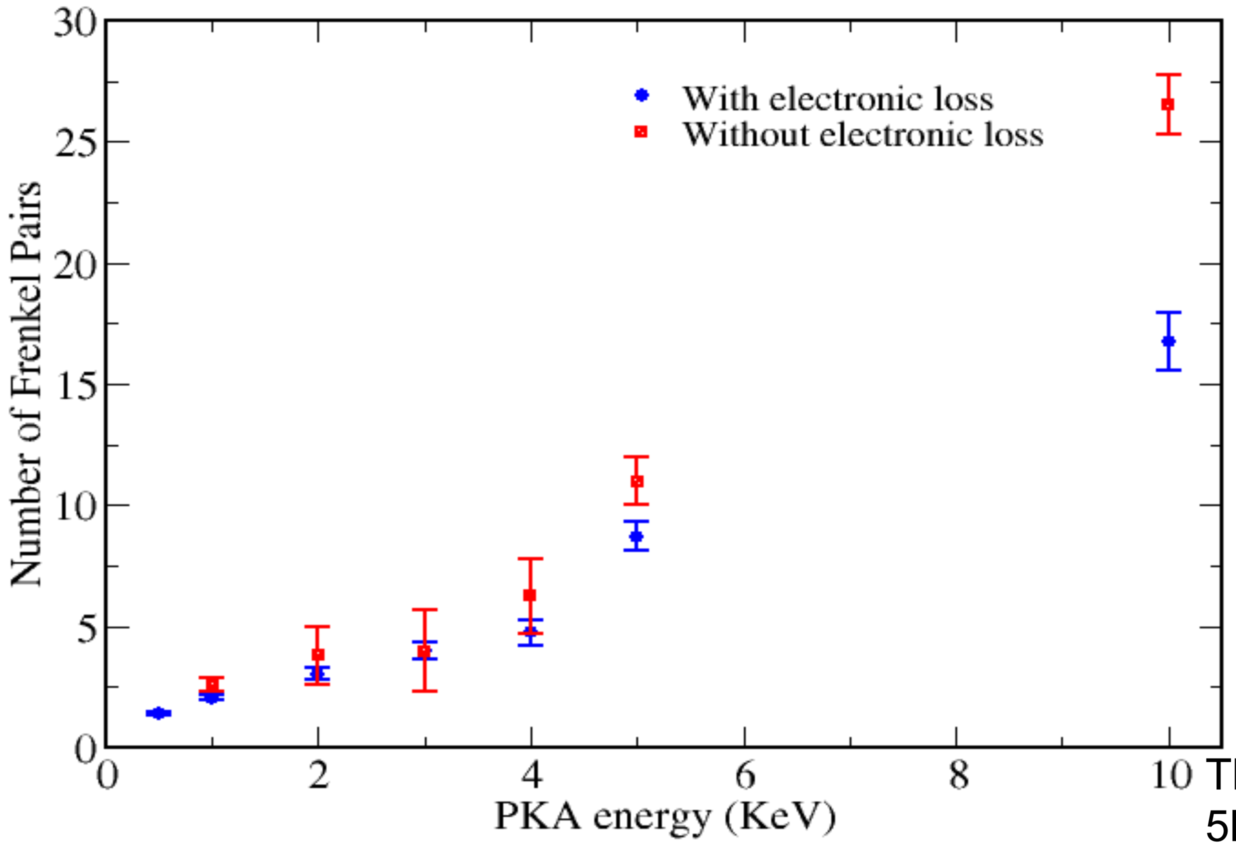


Interstitials in red, Vacancies in green
Atoms participating in the thermal spike at ~1/2 ps have been superposed



Effect of electronic loss in interstitial clustering

Electronic stopping has been taken into account by assuming as a frictional loss to the electron system



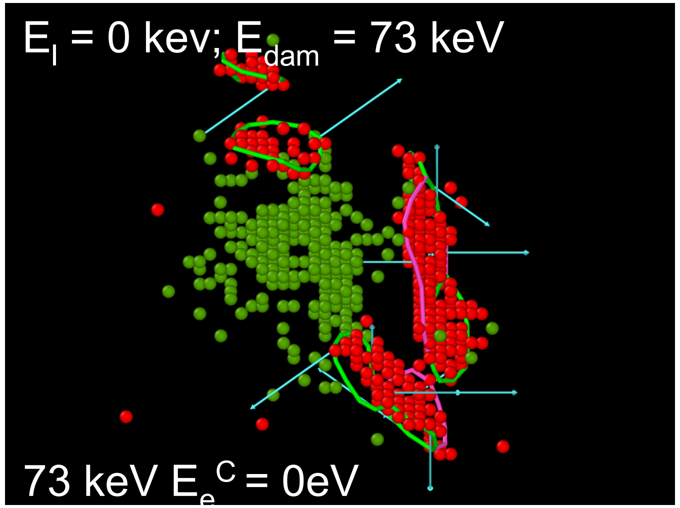
If $v_{ion} < Z1^{2/3}v_B$, energy loss ~ $\sqrt{E}$ ~ v, stopping cross-sections are calculated by Lindhard model

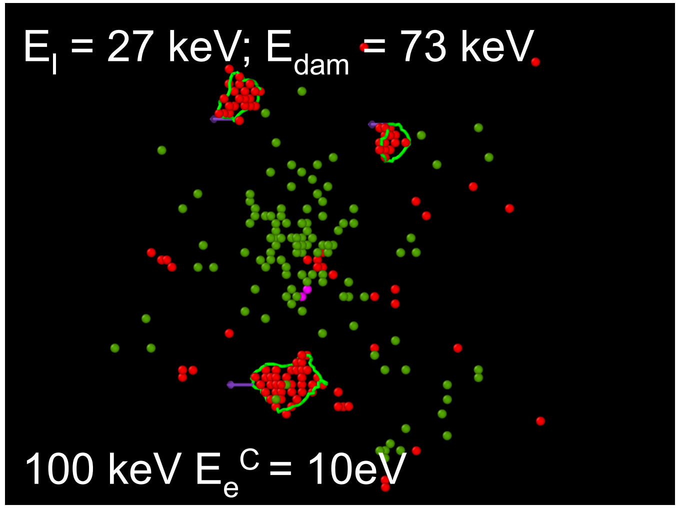
The deviation starts after 5keV

Electronic loss correction at higher energy

- For 100 keV PKA, LSS model yields an electronic loss roughly 27 keV. The energy goes to elastic collisions are hence ~ 73 keV.
- MD simulations are carried out with two conditions: (1) PKA energy with 73 keV and (2) 100 keV with electronic loss such that net damage energy is 73 keV;
- Electronic loss cut-off energy, $E_e^C = 10$ eV.
- It has been observed that the number of defects as well as the structure of defects formed are different.
- Interstitial clustering is observed in both cases. However, the cluster size varied significantly in both the cases for the same PKA directions

Cascade dynamics & NFP changes for the same PKA direction



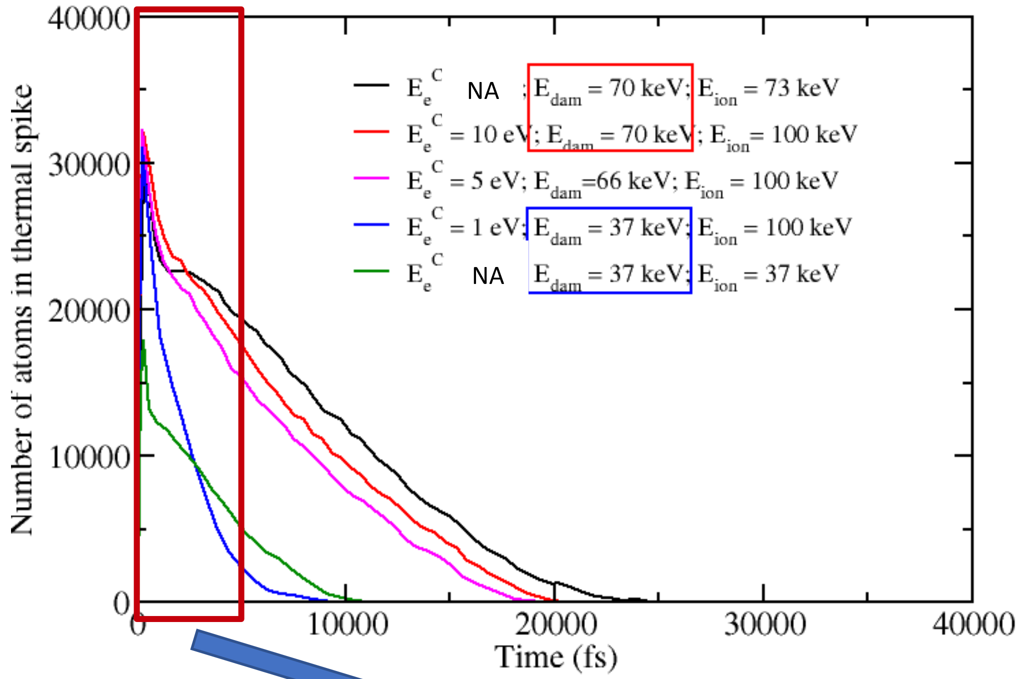


We have also observed similar N_{FP} in some simulations.
→ dynamics of in-cascade clustering depends on the way of energy dissipation of PKA

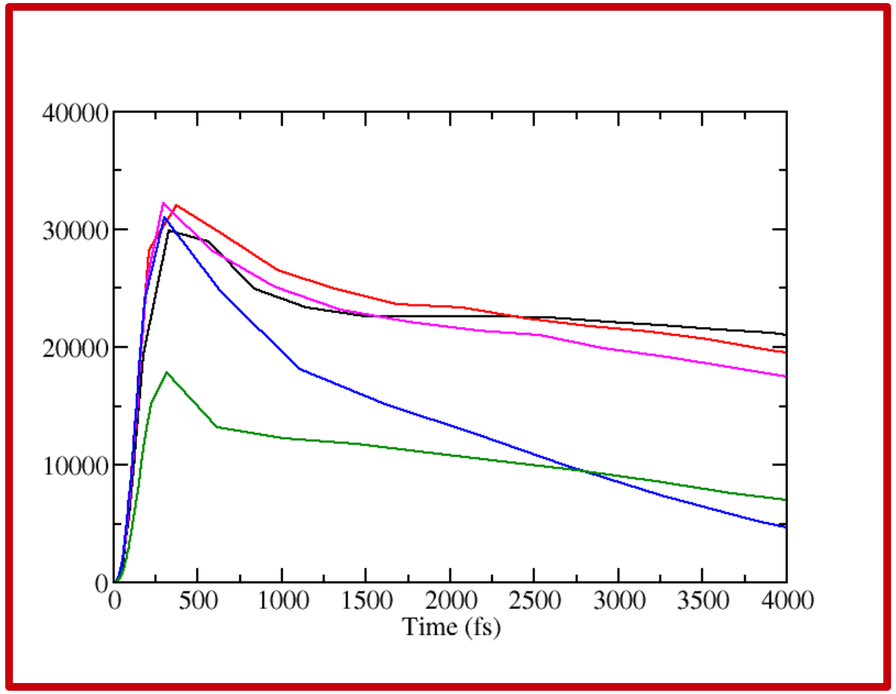
E_{PKA} (keV)	E_e^C (ev)	E_{dam} (keV)	N_{FP}	I_c^{max}	V_c^{max}
100	NA	100	363	154	172
100	1	37	97	28	28
100	5	66	111	54	84
100	10	73	125	44	13
73	NA	73	322	162	117

See also: Sand, Dudarev & Nordlund; Jun 2013 paper, EPL, 103 (2013) 46003

Cascade evolution and thermal spike with different cut-off energies

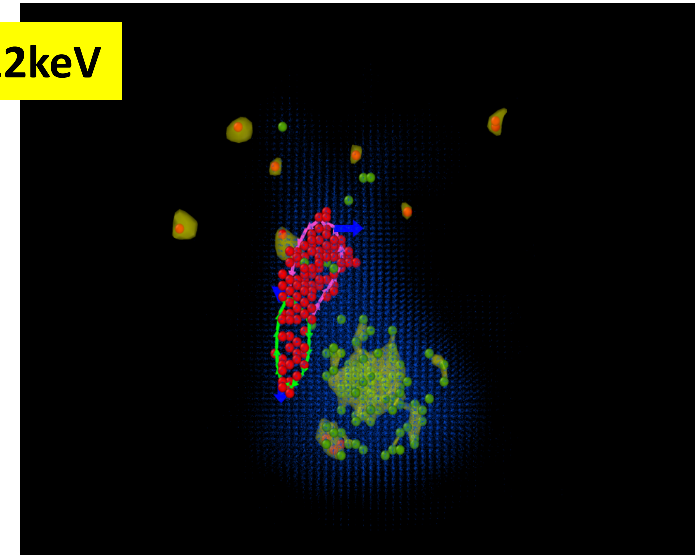


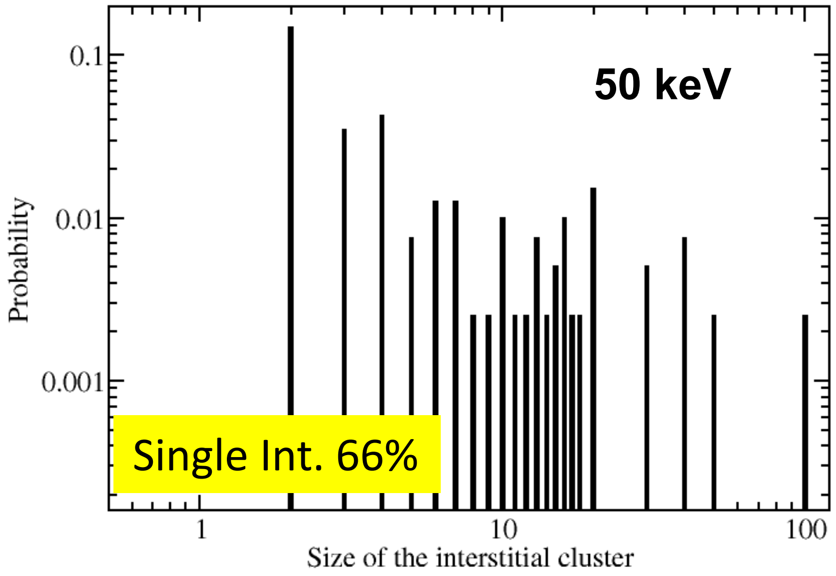
zoom

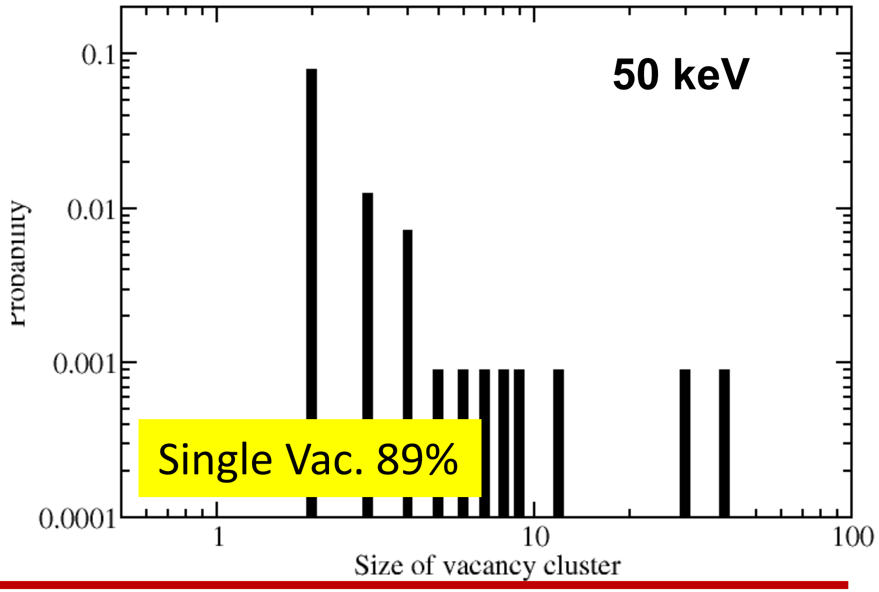


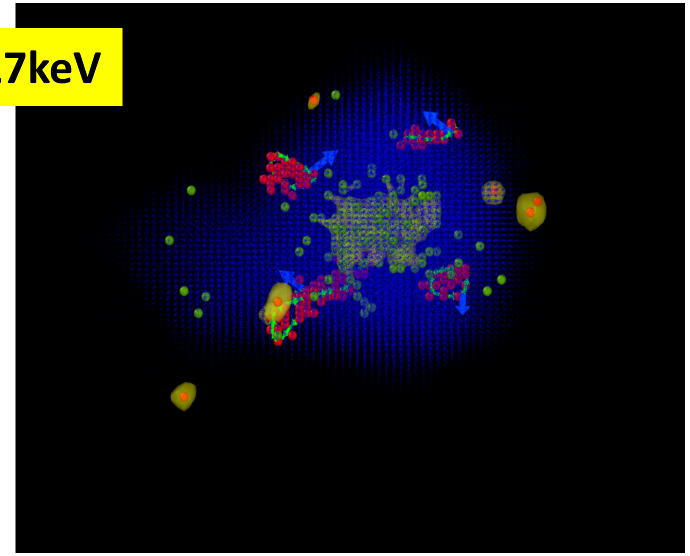
As PKA energy increases larger interstitial & vacancy clusters are formed

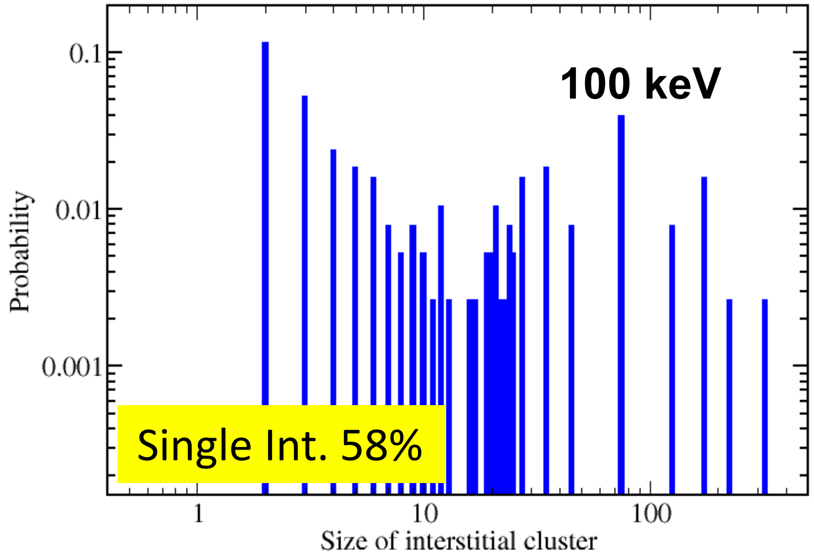
PKA "corrected" for ele. loss

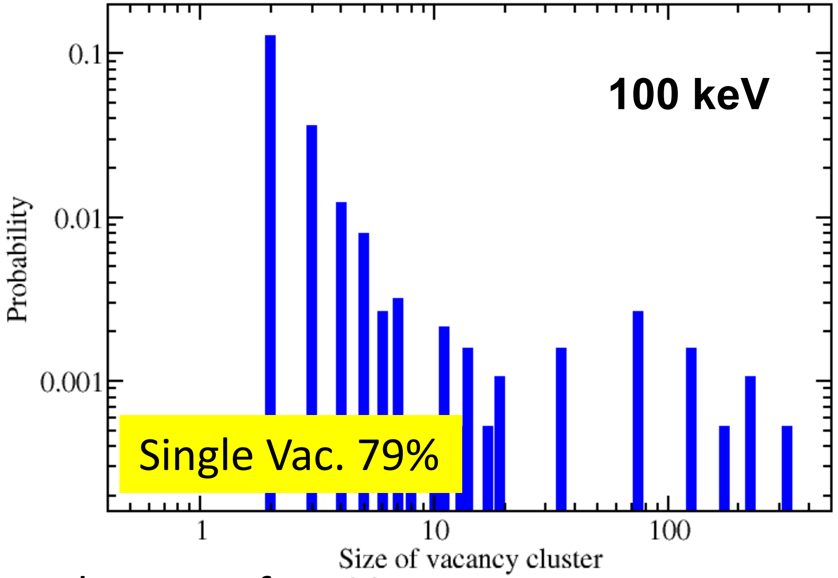




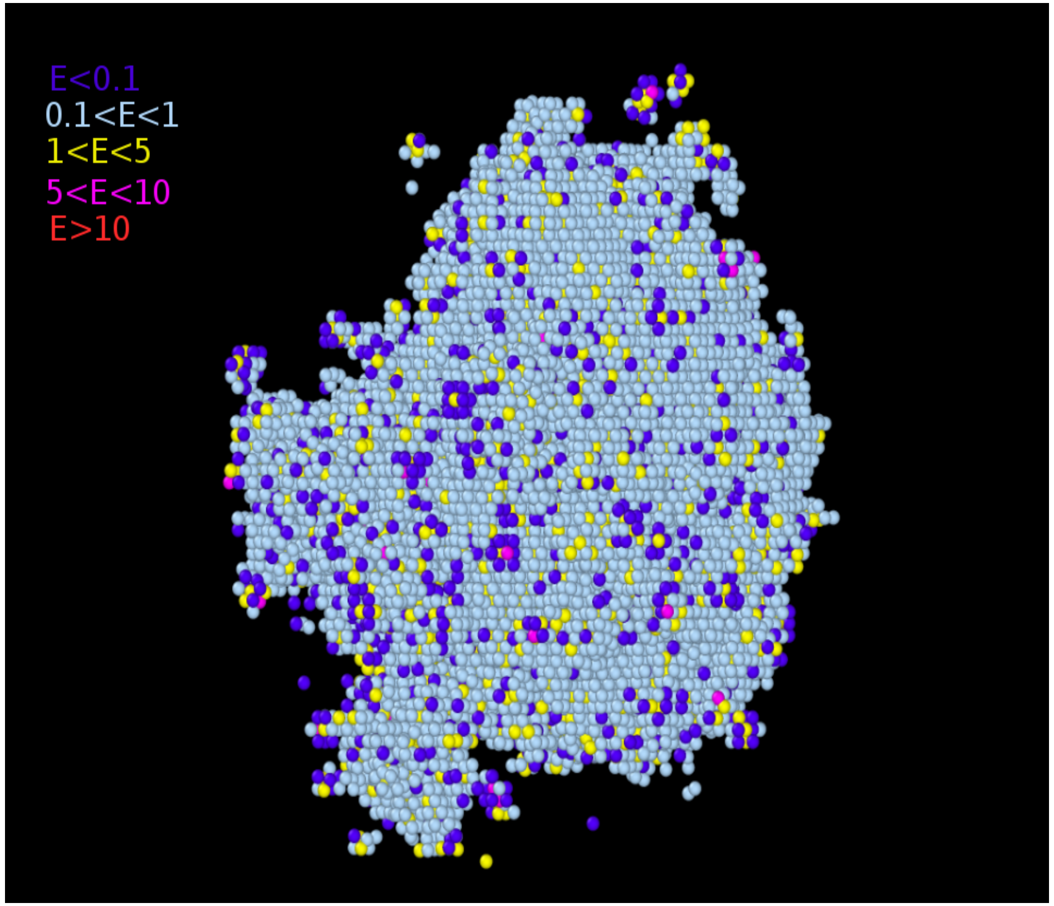




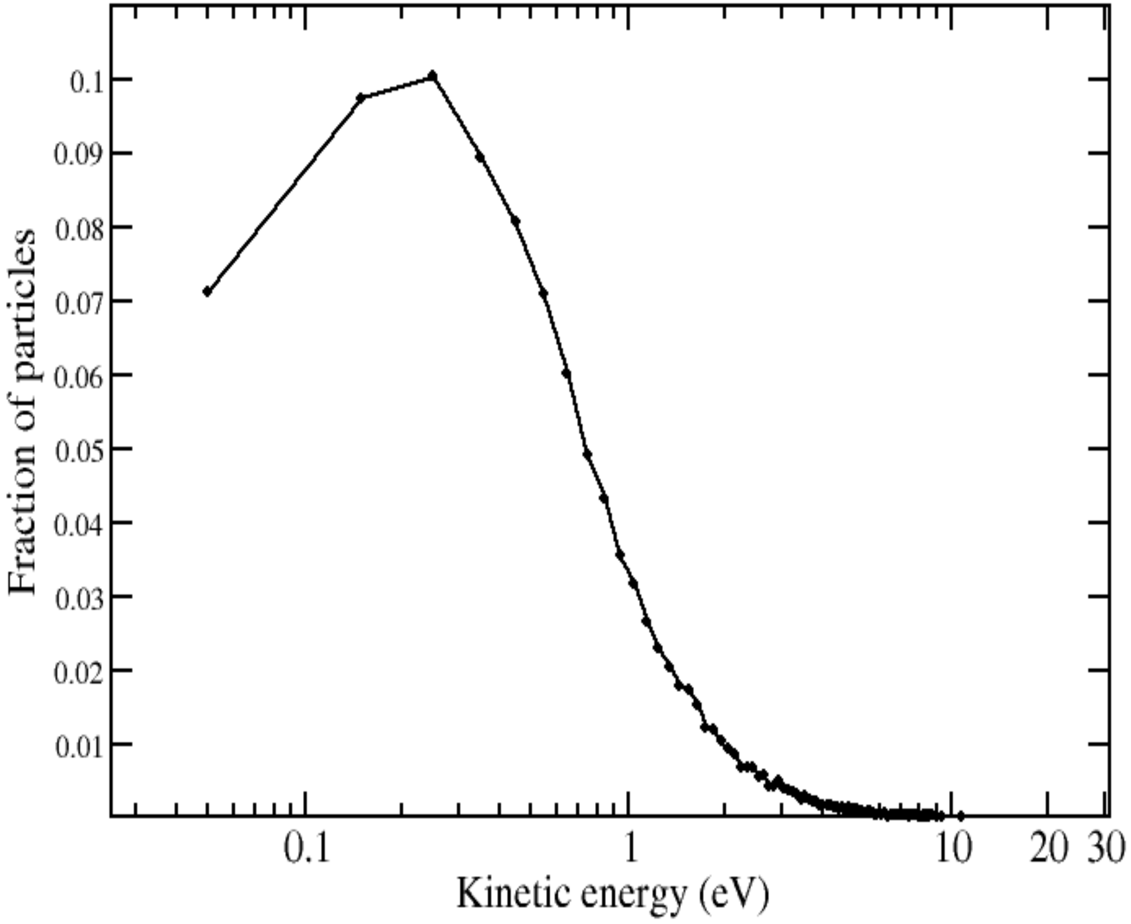




Above plots are after 60 ps



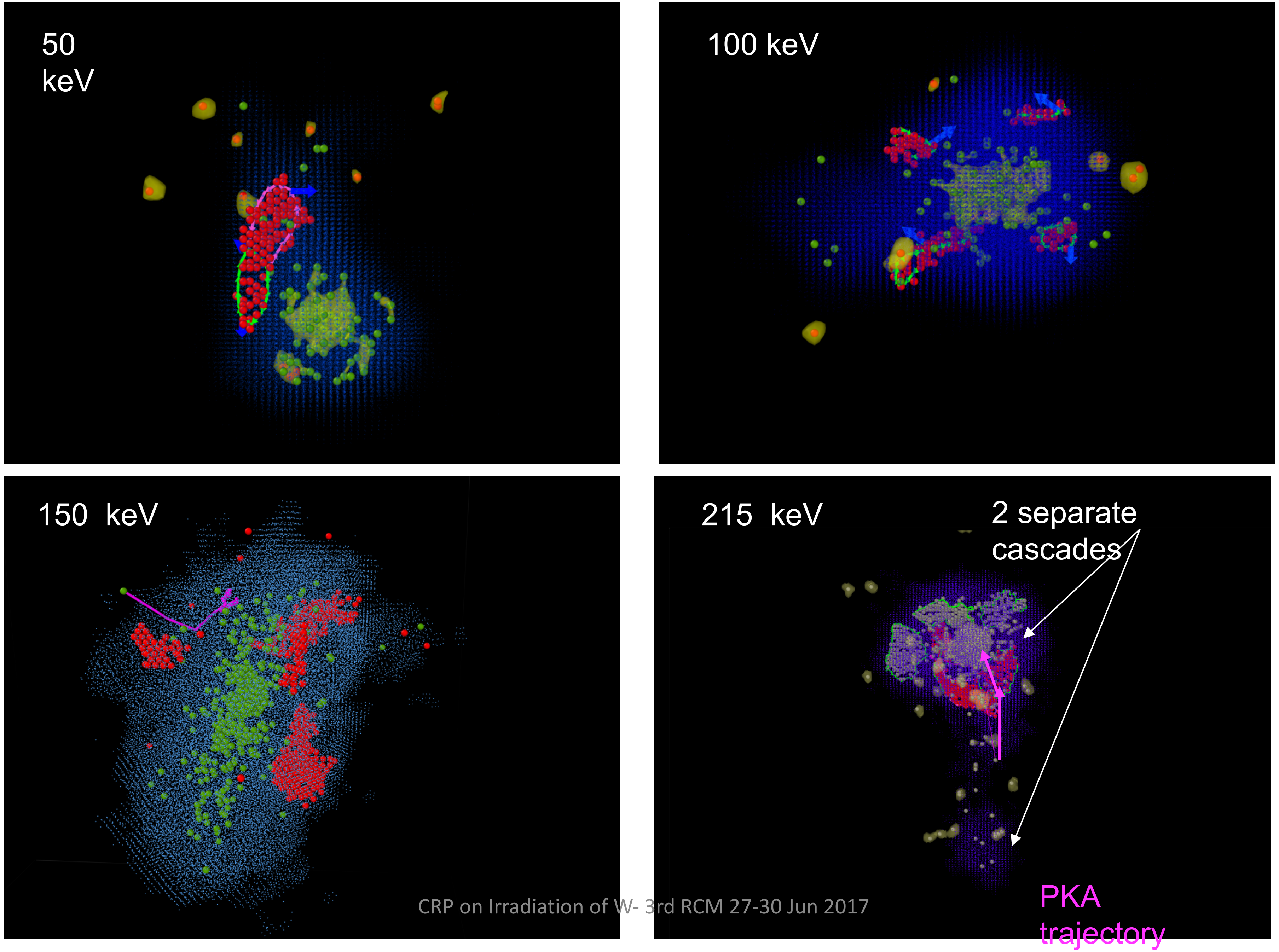
Spatial distribution of KE of interstitials



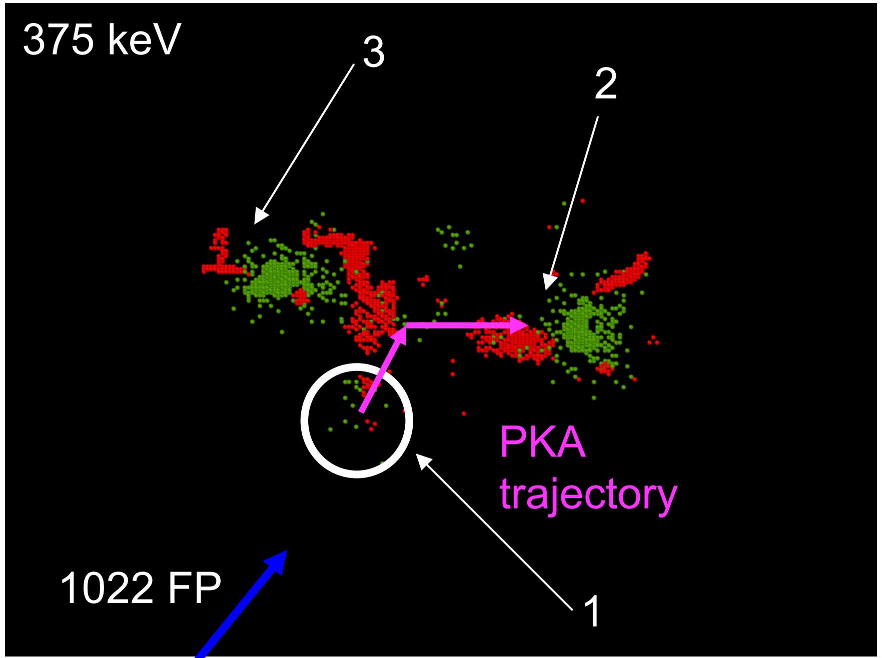
KE distribution of interstitials in the thermal spike

Assuming a solid sphere of radius ~30 interatomic spacings (30x3.16 A) and sharing the KE of 100 keV among the atoms within this radius, one gets roughly 1 eV as the energy per atom; far above the melting temperature

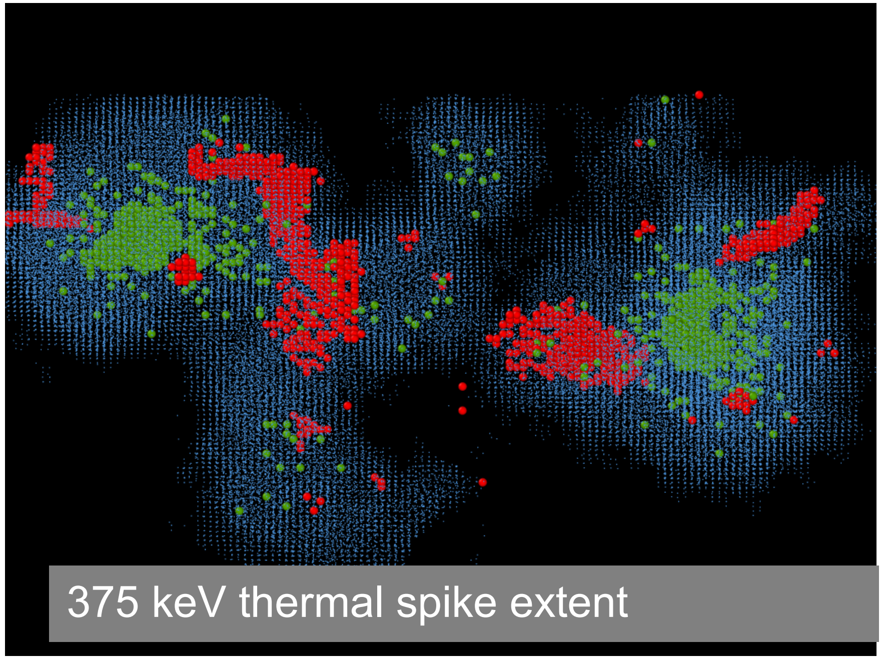
At higher energies ordered clustering of interstitial atoms is observed

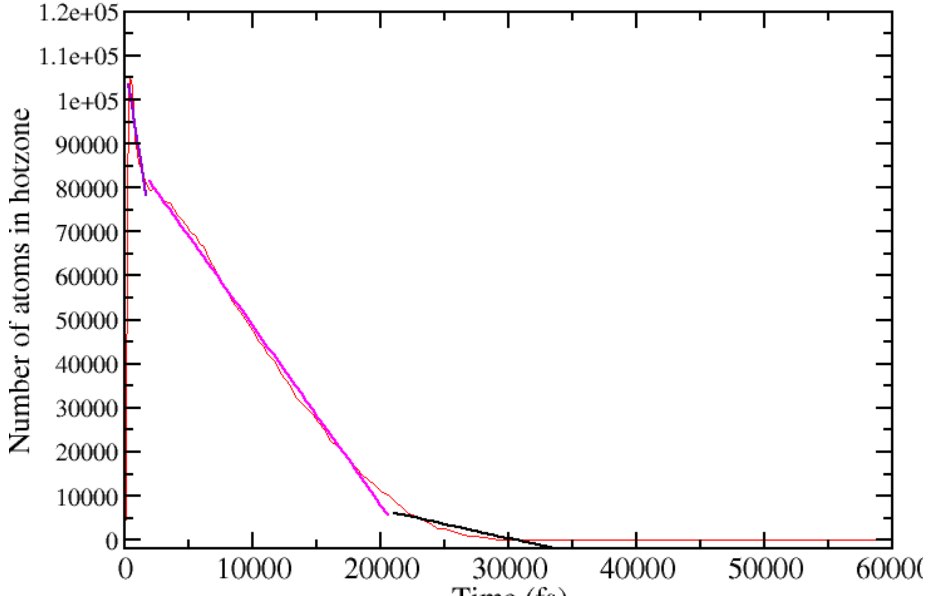


At even higher energies, splitting of cascades observed



100 ps after
the start of
cascade





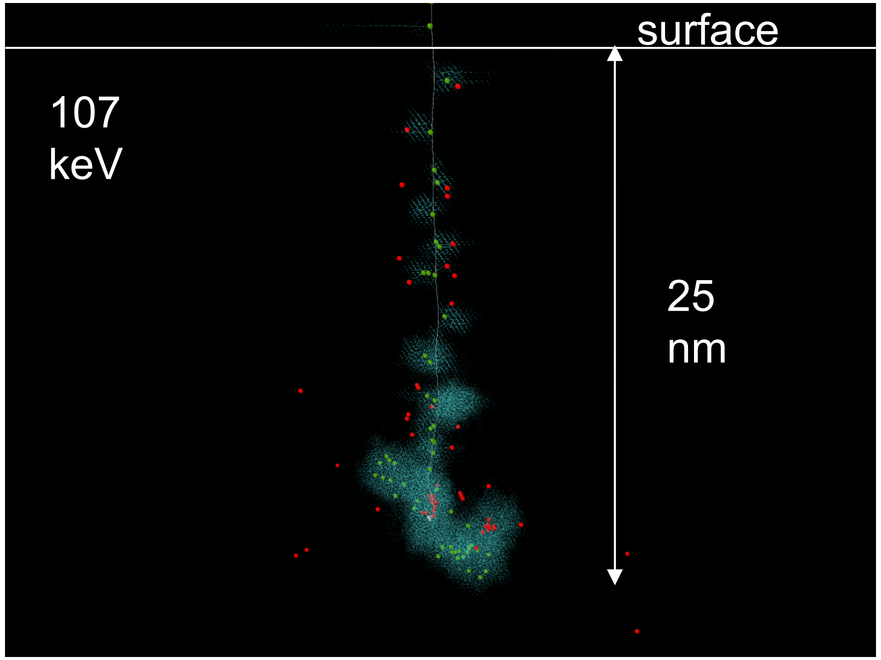
Defect formation near surface

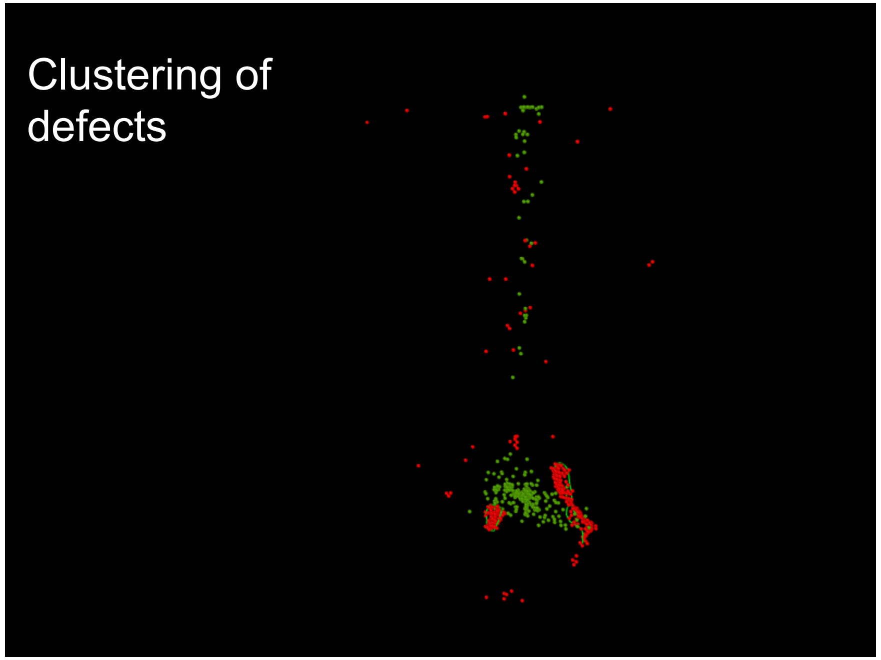
In reality, atoms are bombarded on the surface, hence cascade simulations are carried out with surface

Simulations – 50 keV & 107 keV (to simulate 100 keV Au bombardment on W)

From SRIM: Proj. Range = 11.8 ± 6.3 nm; Sputtering yield = 9.5; electronic loss (LSS) = 21keV; E_{dam} = 86 keV

MD – sample size: 32 nm x 22 nm x 36 nm; mean sputtering yield = 6.6 ± 1.4





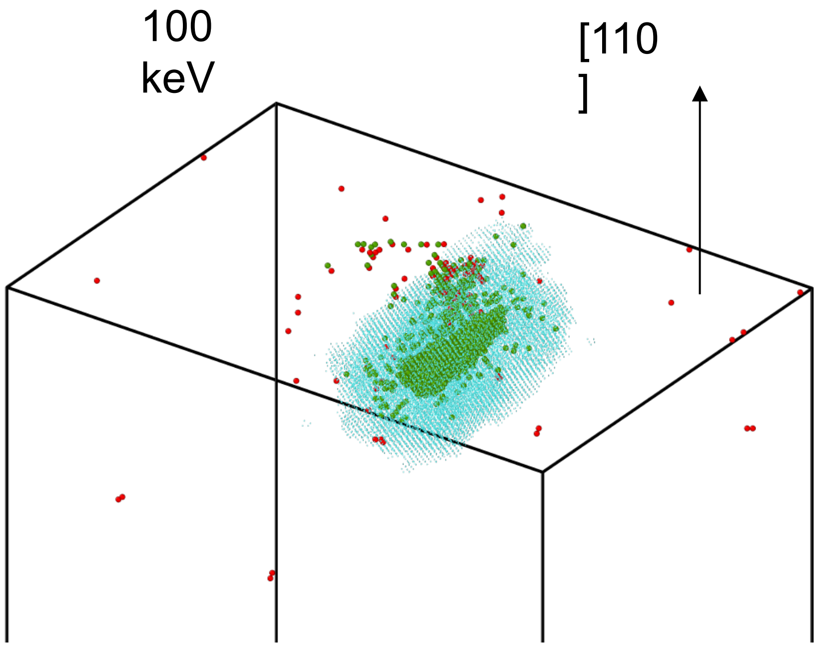
Two types of surface cascades were observed. Scattered thermal spike extended all through out the length of PKA. Some of the cascades leads to clustering of defects.

Average FP obtained in this case was about 45 (from 50 simulation trials).

In a large number of bombardment-events, surface cascades with ordered clustering of vacancies and interstitials were seen. The interstitial clusters were well organized. (21 out of 71 simulations showed this)

Near surface thermal spike

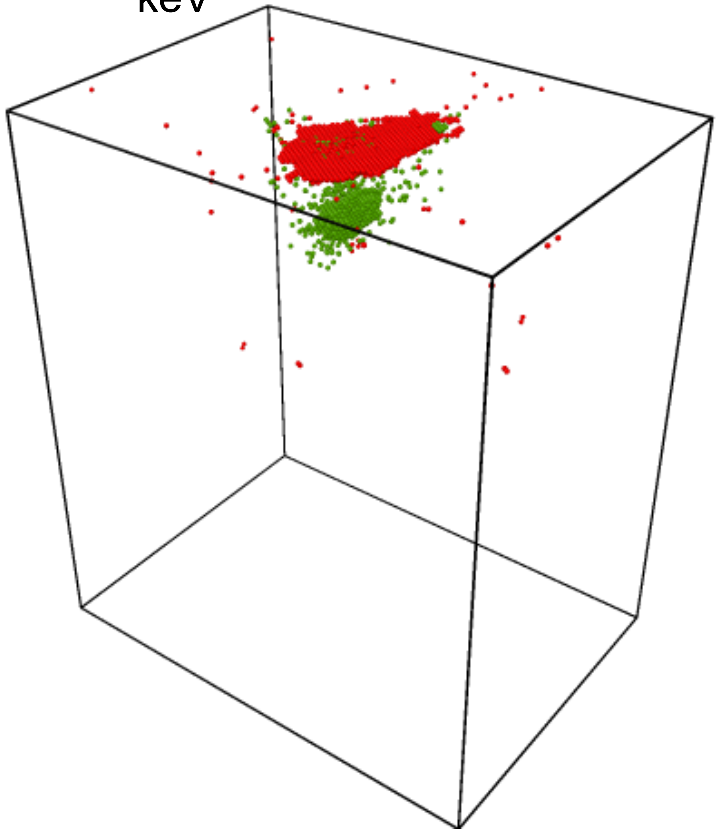
W(110) surface



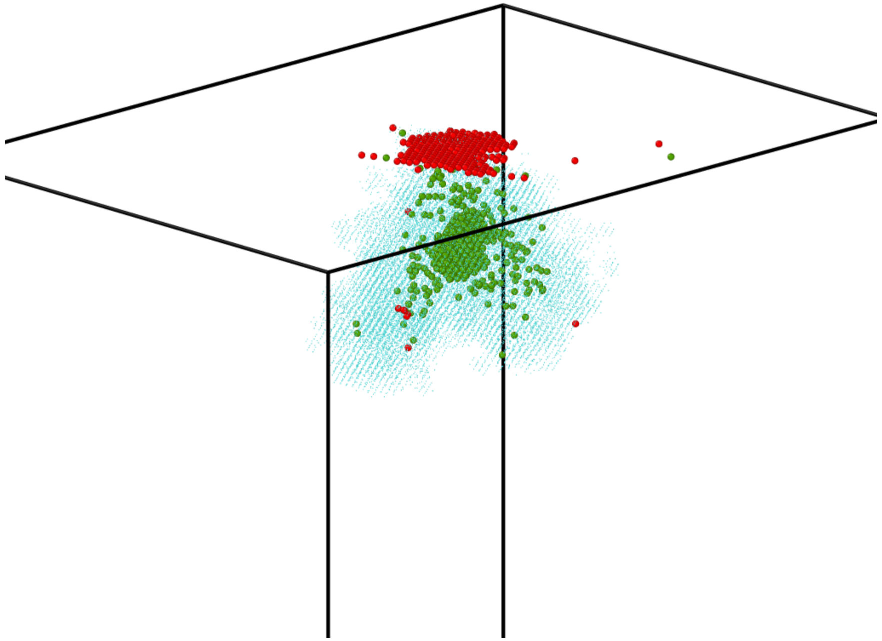
MD simulations have shown that surface cascades can cause stalking faults in FCC metals (Nordlund et.al,., Nature 1999).

Vacancy clustering on the surface may also lead to enhanced erosion as well as crater formation in multiple bombardment.

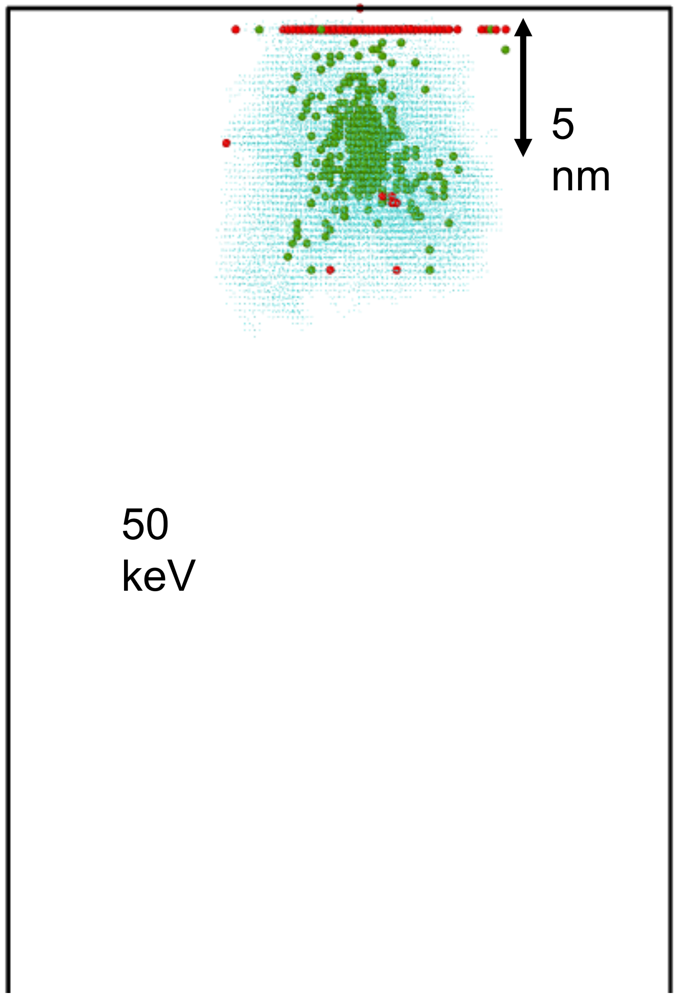
100 keV

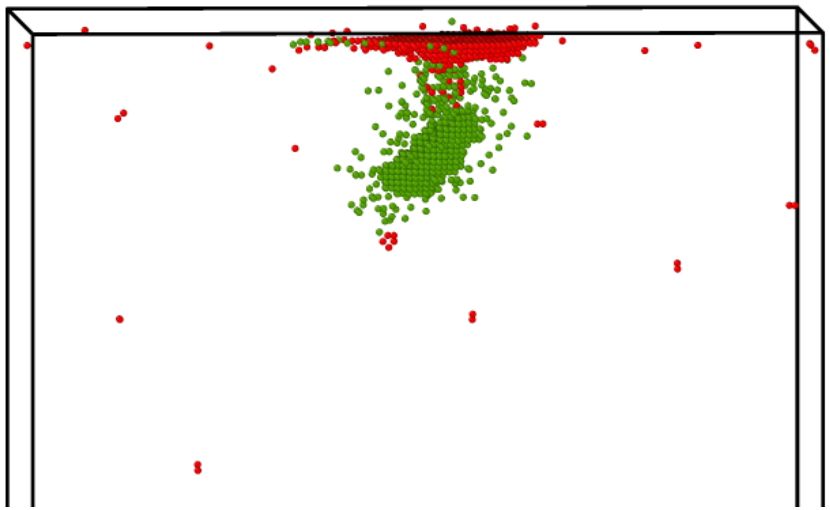


50 keV

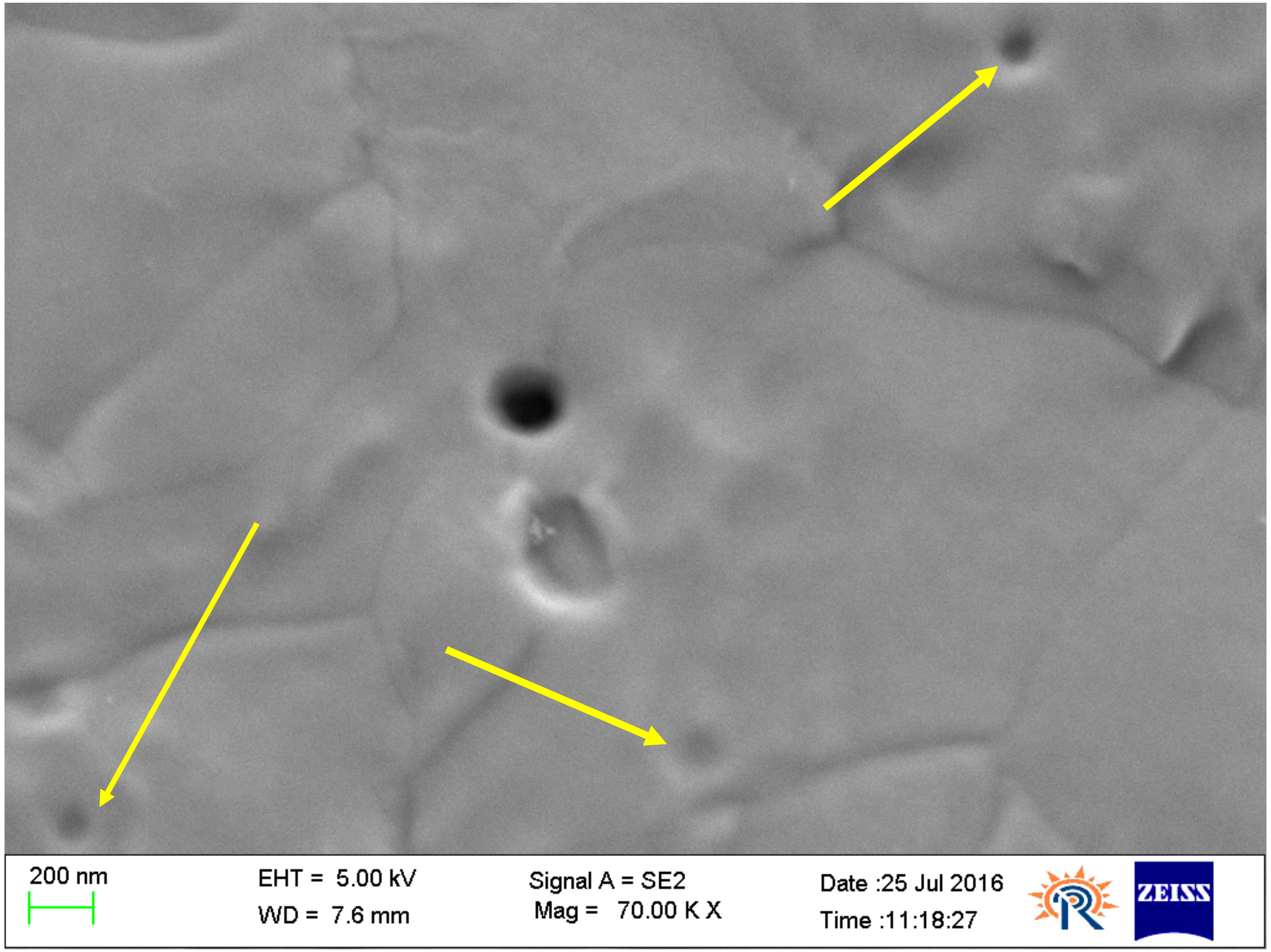


Interstitials and vacancies clustered separately



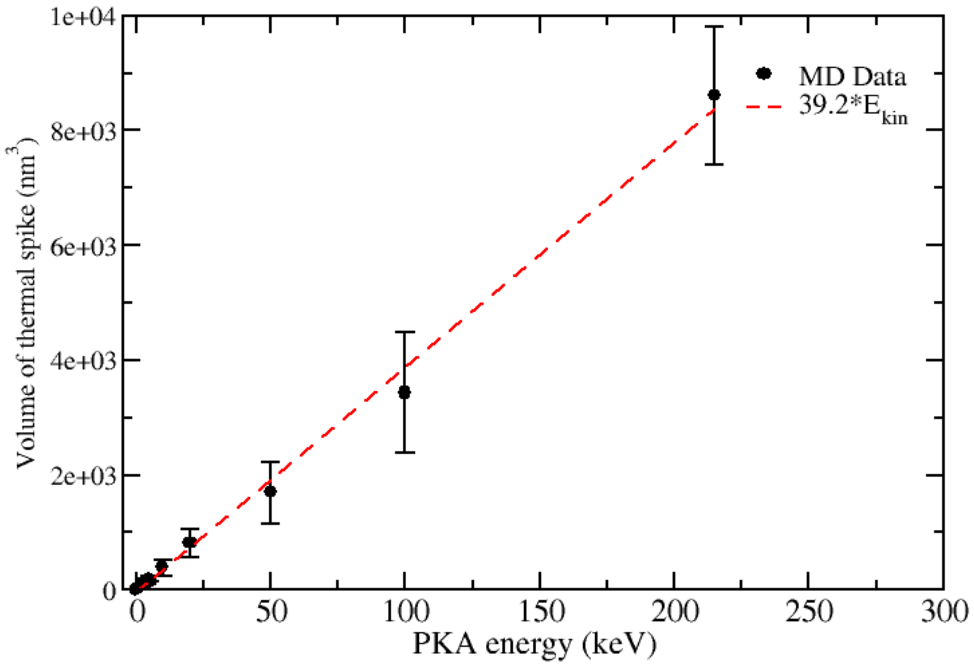


100 keV Au bombardment on plansee W- FESEM images



Formation of large erosion craters on the surface after $1x10^{17}$ ions/cm^2 bombardment on W surface. Some of the pre-existing holes on the sample surface also got enlarged in this process.

Thermal spike volume scales linearly with PKA energy



A rectangular volume of thermal spike is calculated.

Extrapolating to MeV energy regime:

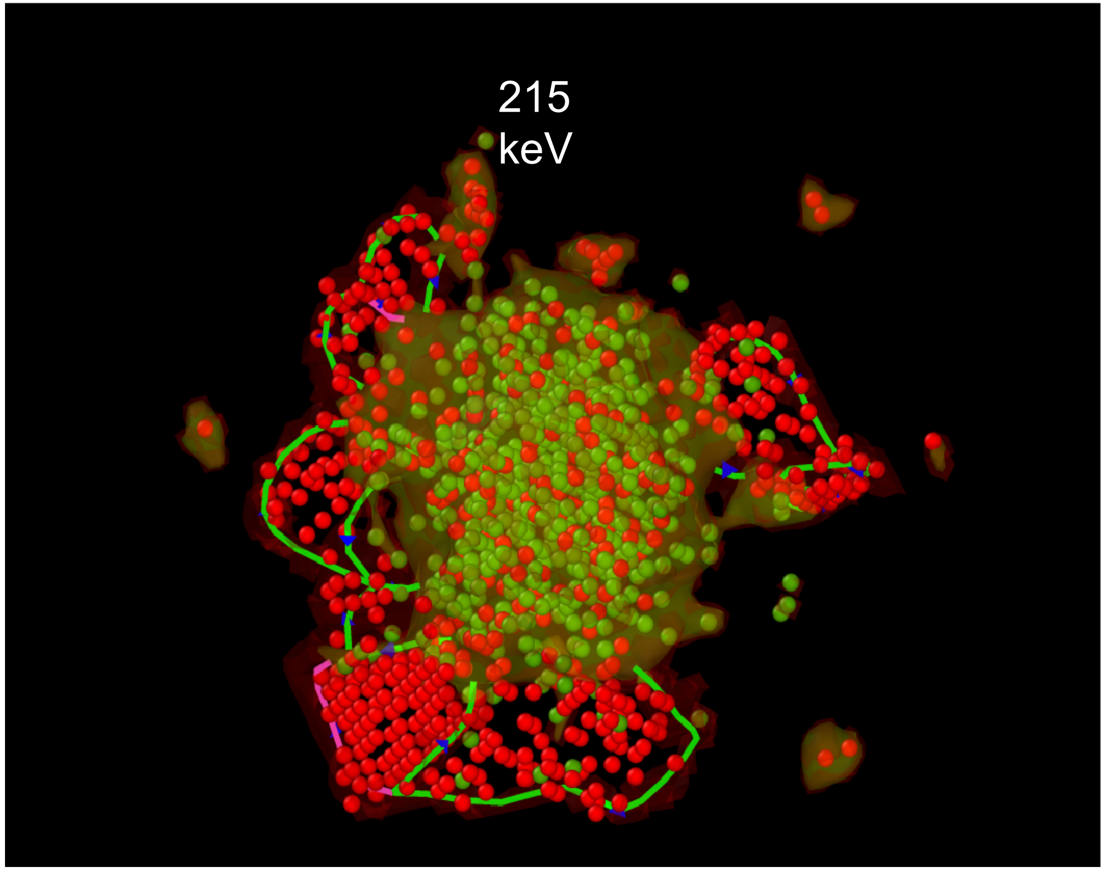
For 80 MeV W-ion bombardment, the thermal spike volume is ~ 3.0×10^6 nm^3 → a crystallite of ~140-150 nm

Cooling down of such a cascade could create multiple dislocations within the crystallite.

Crystallite size estimated from XRD in plansee tungsten: 90 – 140 nm range.

Ion bombardment due to 80 MeV tungsten can cause multiple interstitial clusters within the crystallite

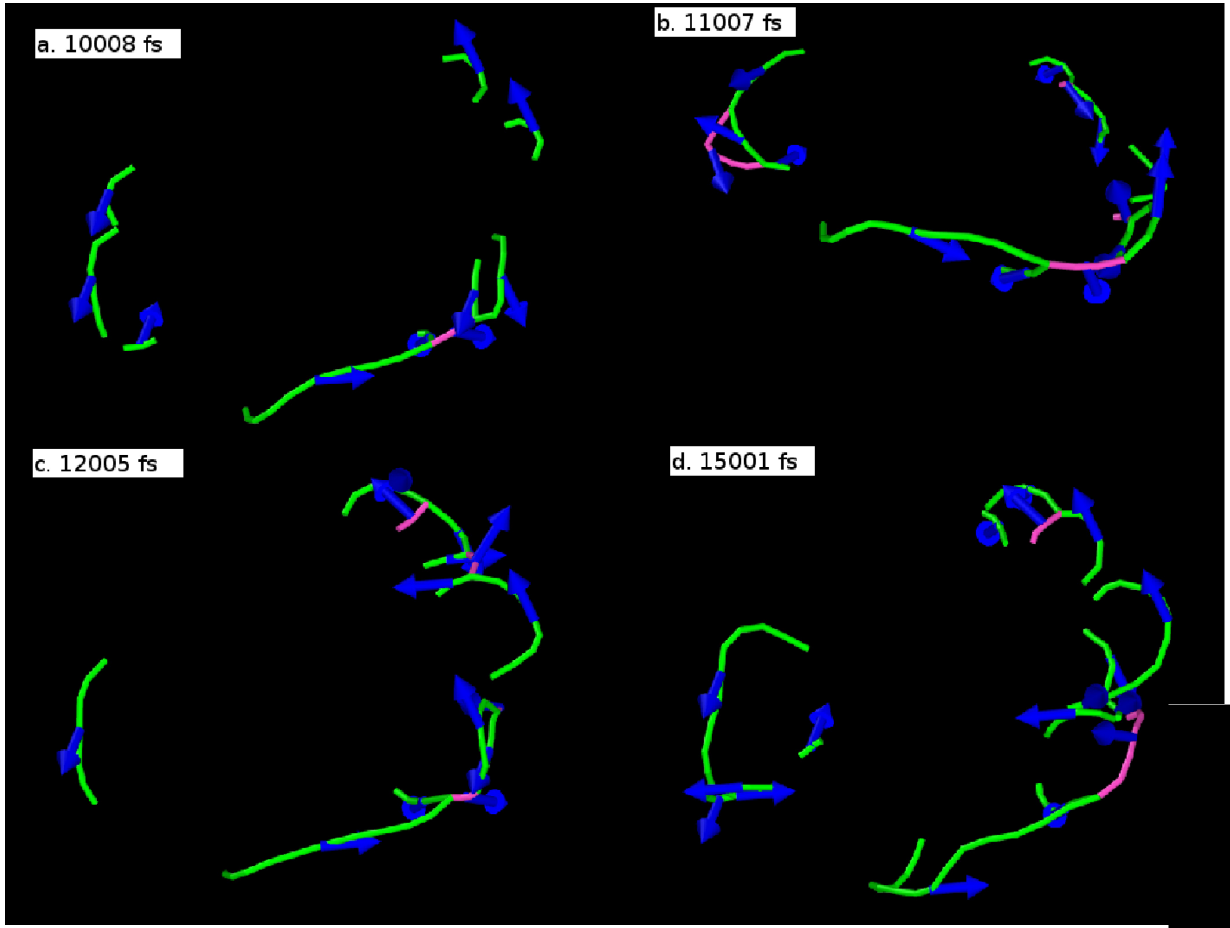
Dislocation analysis carried out using ovito shows that interstitial clustering forms dislocations



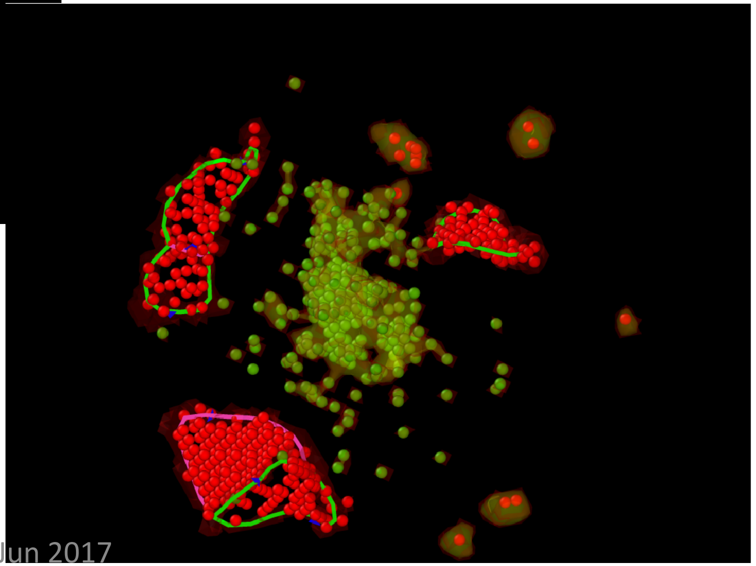
Both <100> and 1/2<111> type dislocations were observed in the simulations

<100> arrangement of atoms have been observed by merging of two 1/2<111> dislocated atoms

Short Time Scale Interactions of Dislocations



After 100 ps of bombardment



We have not observed any vacancy dislocations in the timescale of simulations

Experiments

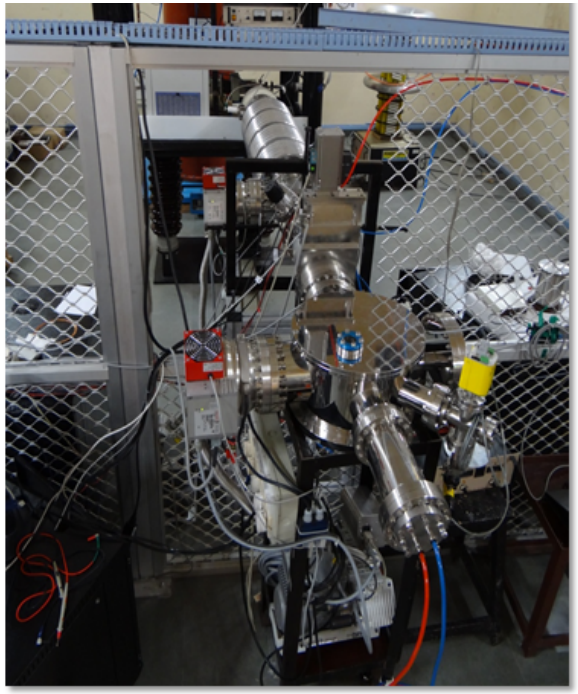
Experiments Conducted

- Irradiation Experiments
  - 80 MeV Au^{7+} on recrystallized W-foils (8mmx8mmx100μ)
  - 80 MeV Au^{7+} on W-La_2O_3 alloys (8x8x1mm^3)
  - 10 MeV B^{3+} on recrystallized W-foils (8mmx8mmx100μ)
  - 250 keV He^{+} on recrystallized W-foils (8mmx8mmx100μ)
  - 100 keV D^{+} on recrystallized W-foils (8mmx8mmx100μ)

- Characterization
  - Defect Analysis - PAS, TEM, Resistivity
  - Microstructural Studies - XRD, FESEM, TEM
  - Deuterium depth profile - SIMS

Experimental Facilities

Facility	Ion used	Energy
15 MV Pelletron, IUAC Delhi	W, Au	80 MeV
3 MV tandem accelerator, GGU, Bilaspur	B	10 MeV
Low energy metal ion implanter, IUAC Delhi	Au	100 keV
Low energy gaseous ion implanter (LEIBF), IUAC Delhi	He	250 keV
Accelerator based 14 MeV neutron generator, IPR	D	100 keV



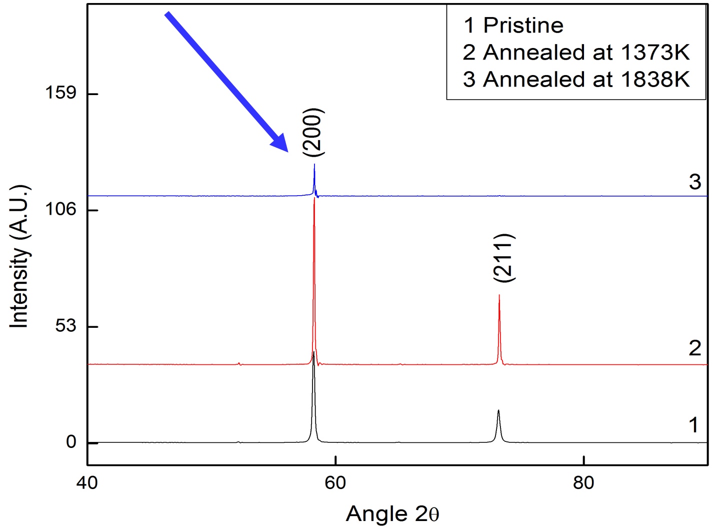
Accelerator at IPR for 100 keV D implantation

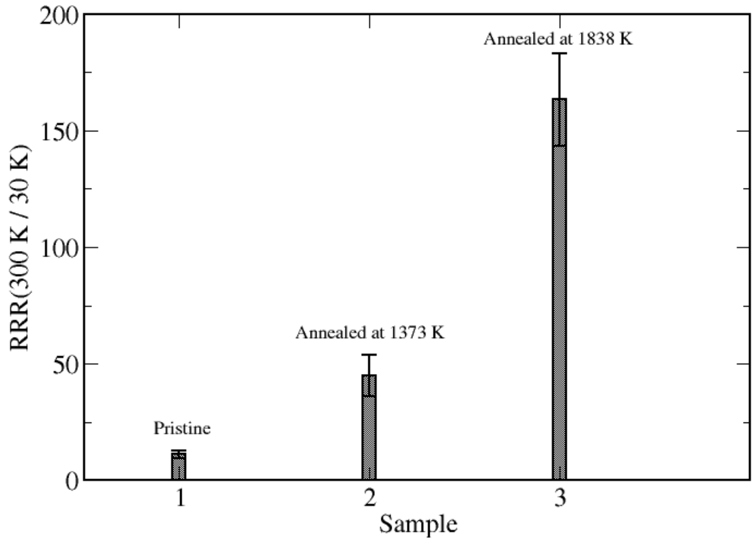
3 MV Accelerator at GG University,Bilaspur,India used for 10 MeV Boron irradiation

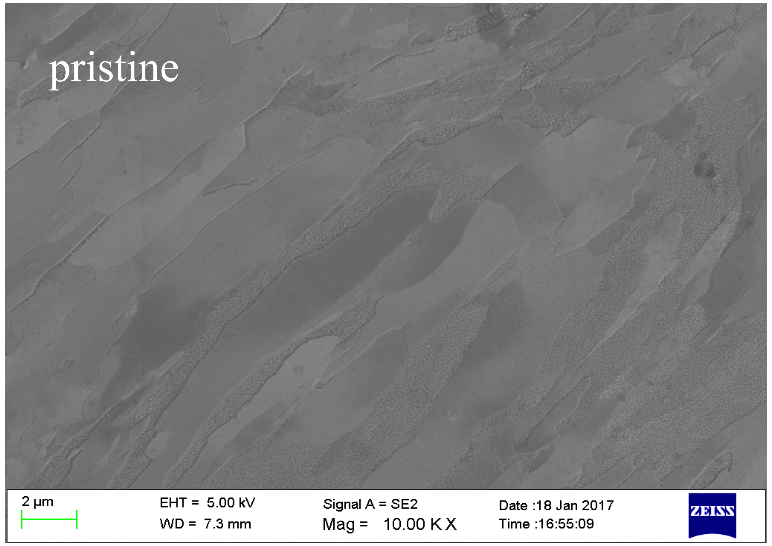
Samples Used for Experiments

Tungsten foils of 100 μ thickness were procured from Princeton scientific corp, USA

Annealing temperature: 1373 K & 1838 K; Vacuum = 10^{-4} mbar with 100 mbar Ar + 8 % H; time – 50 min

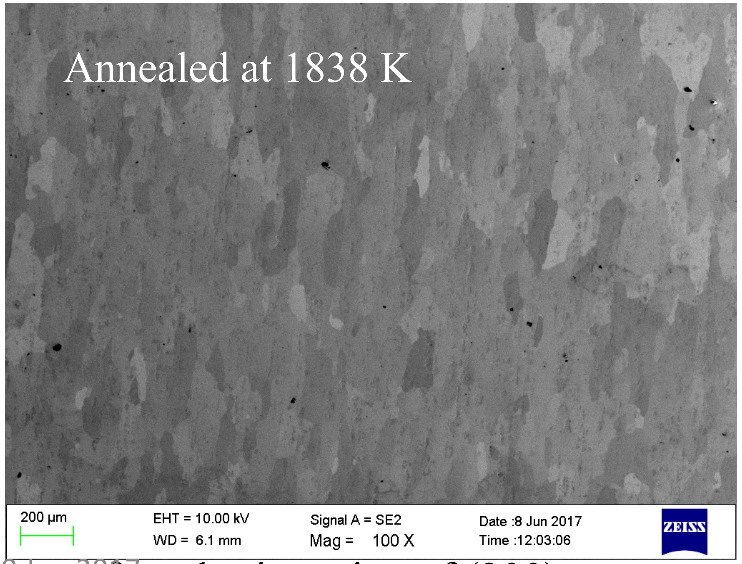




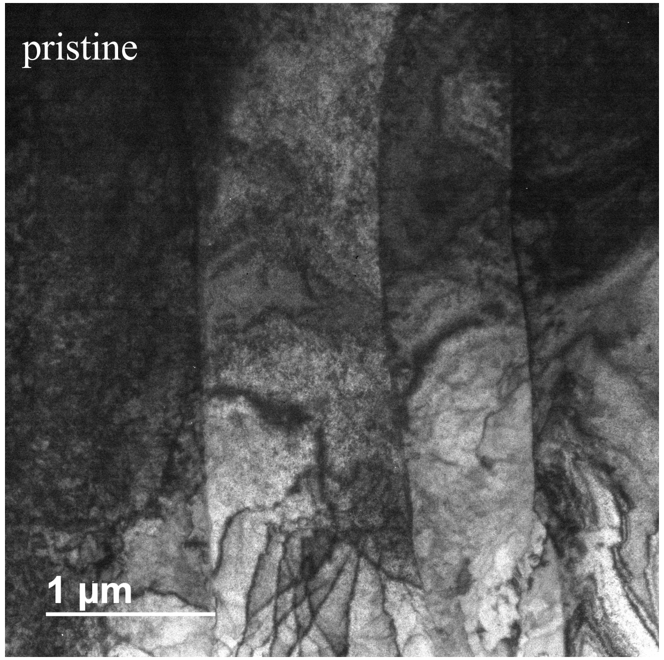


Average crystallite size from XRD

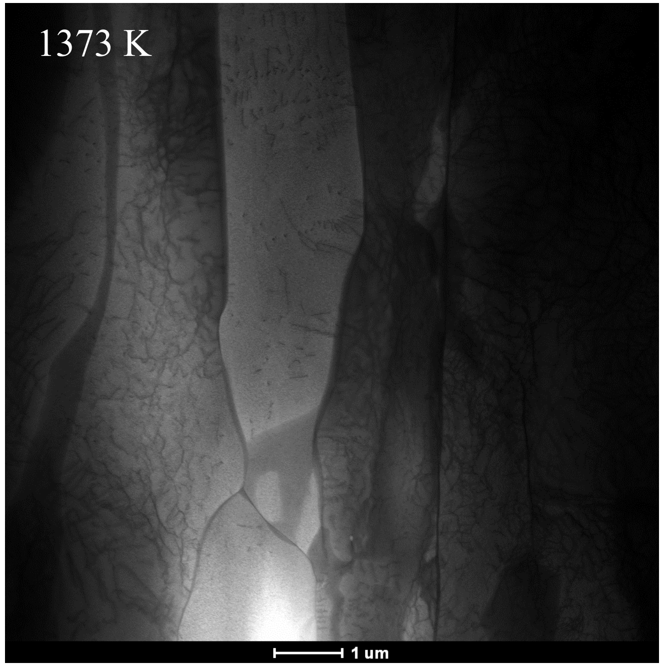
Pristine ~ 64 nm
1373 K ~ 300 nm
1838 K > 700 nm



After 1838 K annealing the samples were recrystallized with a preferred orientation of (200)

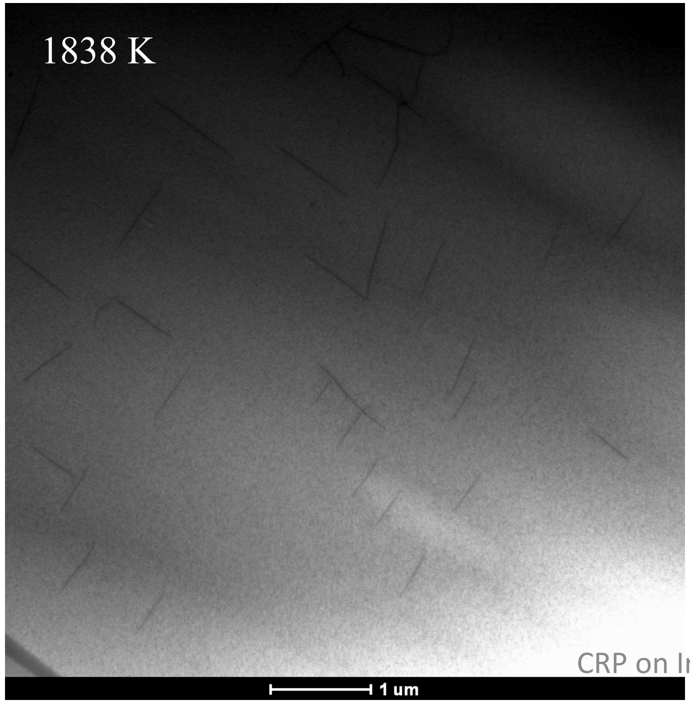


TEM micrographs showing dislocation annealing and grain growth with annealing temperature



Dislocation line length = [200 nm to 900 nm]

Dislocation density = 3.6 x 10^{8} cm^{-2}



Point defect annealing:
from positron life-time data for 1838 K

τ_1 = 107.3 ± 1.7 ps (70 % intensity);

(105 ± 3 ps for perfect W lattice with ~100 % intensity)
Ref: Troev et.al., JPCS, 207 (2010) 012033

τ_2 = 237.3 ± 5.6 ps (30% intensity); → vacancy clusters

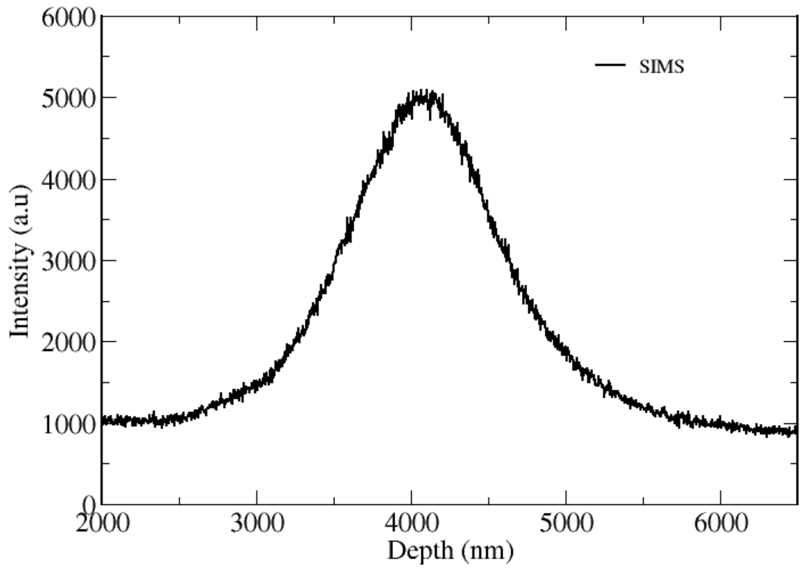
All Irradiation experiments were carried out on recrystallized foils

High Energy Heavy ion Irradiation

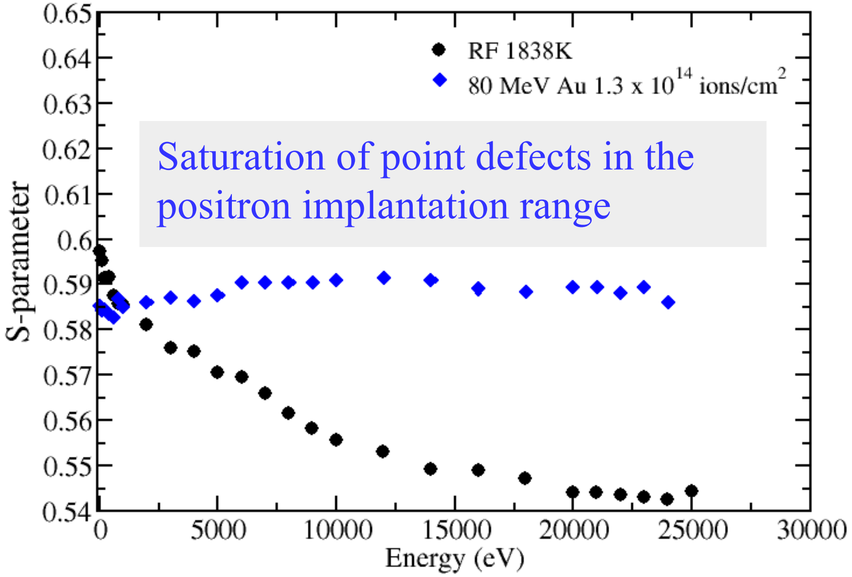
Ion →Au $^{7+}$; Energy → 80 MeV; Flux → 9.76 x 10^9 ions/cm^2/s; Fluence → 1.3 x 10^{14} ions/cm^2
dpa = 0.22 for E_d = 90 eV

Au implantation profile

$R^{SIMS}_{mean} = 4.06 \pm 0.5\ \mu m$
$R^{SRIM}_{mean} = 4.49 \pm 0.73\ \mu m$

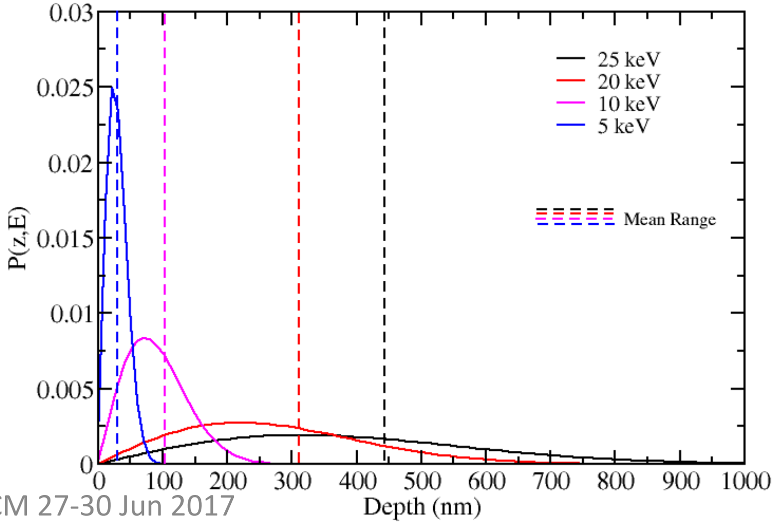


Positron Doppler broadening



Similar observations of defect saturation for 0.022 dpa, reported by MF Barthe, CEMHTI CNRS Orléans, MINOS , Cadarache 4-6/11/2015

Mean e$^+$ implantation depth for various energies by assuming Makhovian profile



Defect Analysis

Bulk positron life-time using [22]Na source with mean penetration range of 15 μm

τ_1 = 130.4 ± 4.9 (intensity → 52 ± 4)
τ_2 = 250.5 ± 7.5 (intensity →45 ± 4)

Splitting into the recrystallized foil life times,

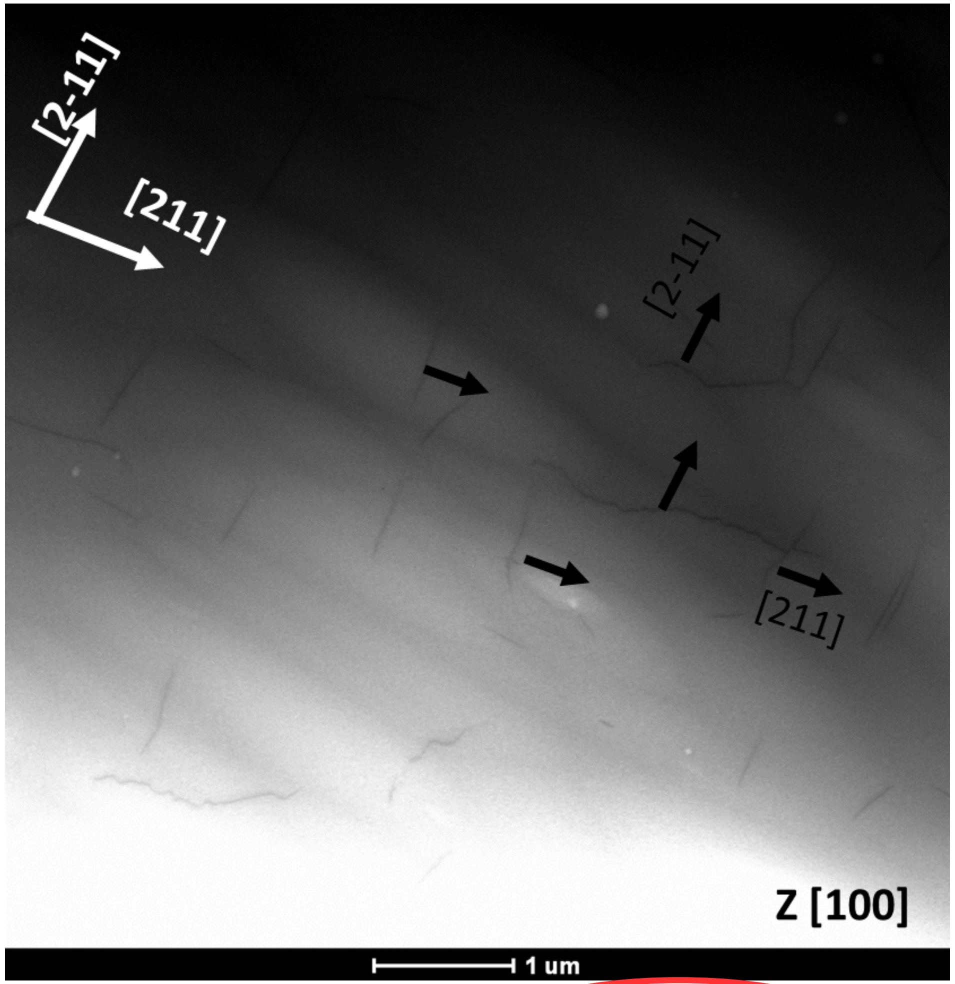
τ_1^{RF} = 107.3 (18 ± 3) ; (down from 70 %)

τ_2 = 154.0 (41 ± 7); → *positron annihilation from dislocations*

τ_3 = 260 ± 9 (37 ± 3) → vacancy clusters

Long dislocation lines ~ [0.4 μm to 2 μm]

STEM image (Au Irradiated)



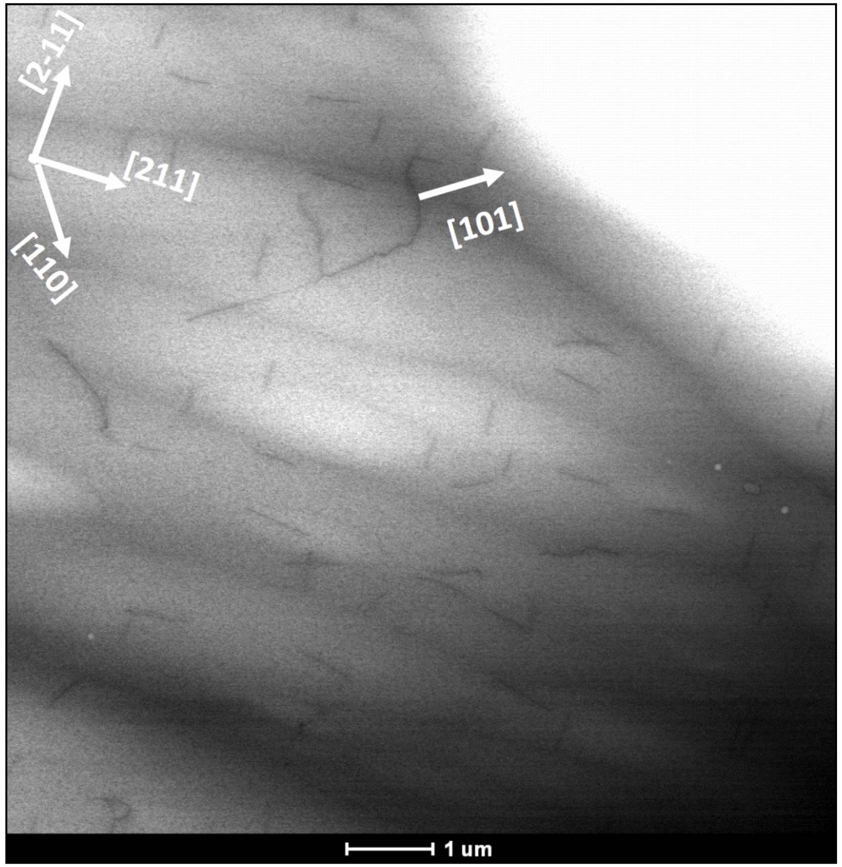
Dislocation density = 2.9 x 10^8 cm^{-2}

For unirradited foil = 3.6 x 10^8 cm^{-2}

Dislocations in depth

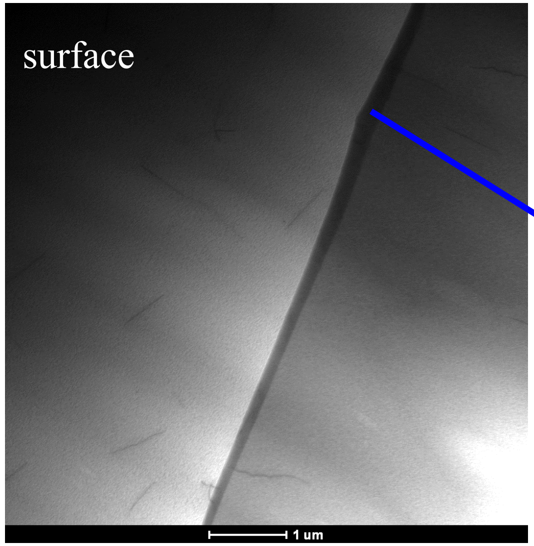
Average dislocation length = 0.4 µm to 3 µm
Dislocation density = 6.1 x 10^8 cm^{-2}

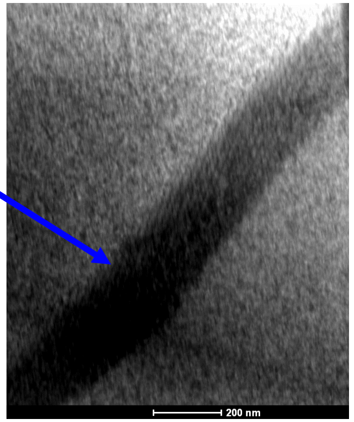
2 µm in depth

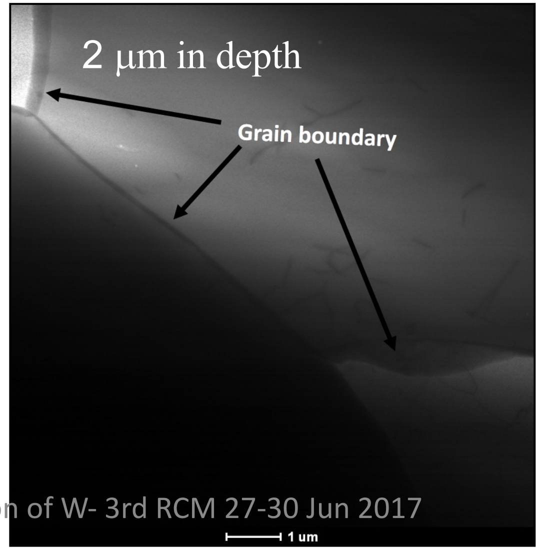


No loops were seen within the measurement zone

Grain boundary





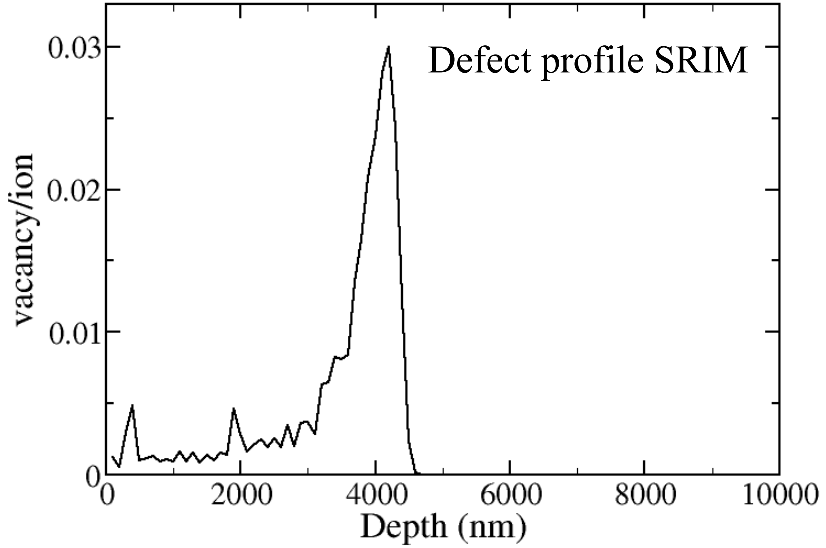


Accumulation of defects at the grain boundary (??)
→ Widening of GB

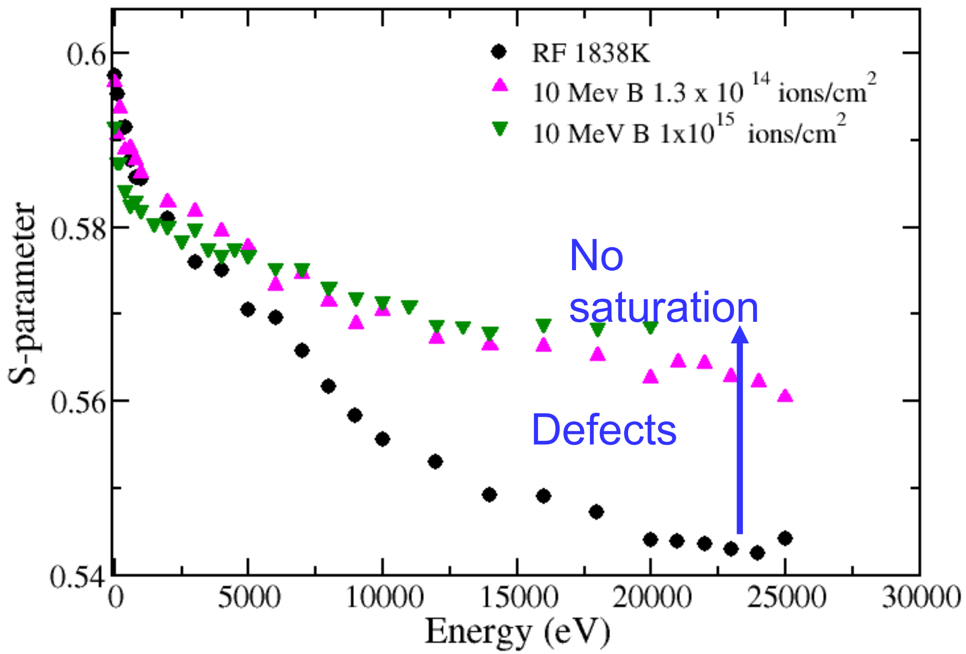
High Energy Light ion Irradiation

Ion →B $^{3+}$; Energy → 10 MeV; Flux → 9.8 x 10^{10} ions/cm^2/s; Fluence → 1.3 x 10^{14} ions/cm^2 and 1 x 10^{15} ions/cm^2; dpa = 0.001 and 0.01 for E_d = 90 eV

R^{SRIM}_{mean} = 4.1 ± 0.3 μm; similar to 80 MeV Au range (4.49 ± 0.73 μm)



Positron DB measurements

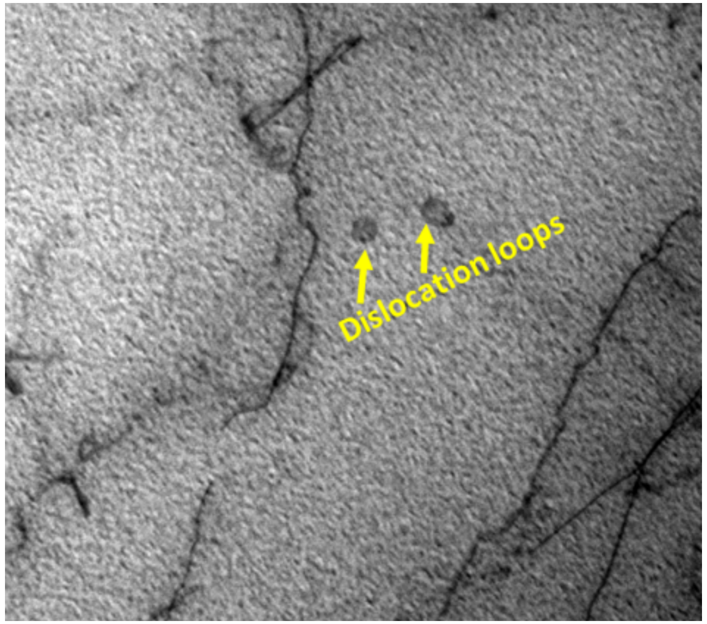


Fluence (ions/cm^2)	τ_1 (ps) (Intensity)	τ_2 (ps) (Intensity)
0	107.3 ± 1.7 (70 %)	237.3 ± 5.6 (30%)
1.3 x 10^{14}	117.1 ± 2.4 (58 %)	232 ± 4.6 (38 %)
1x 10^{15}	117.7 ± 2.6 (50 %)	219 ± 3.5 (46 %)

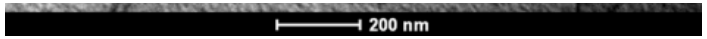
Positron bulk life-time measurements

Small variation in defect concentration with fluence

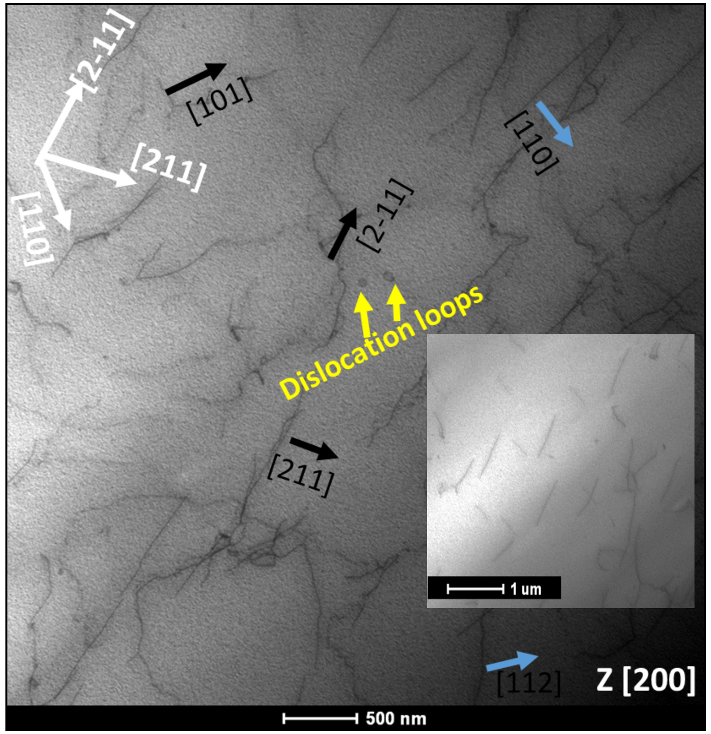
Dislocation loops & smaller dislocation length in B irradiated Tungsten

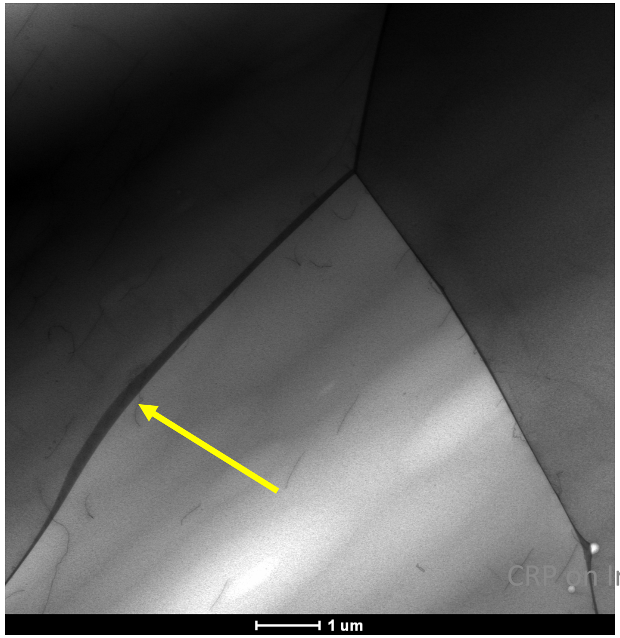


Loops are found closer to larger dislocations



Boron fluence:
1x 10^{15} ions/cm^2





Dislocation density: **7.9 x 10^{8} cm^{-2}**

higher than 80 MeV Au irradiated!

Dislocation line length: **50 nm - 900 nm**

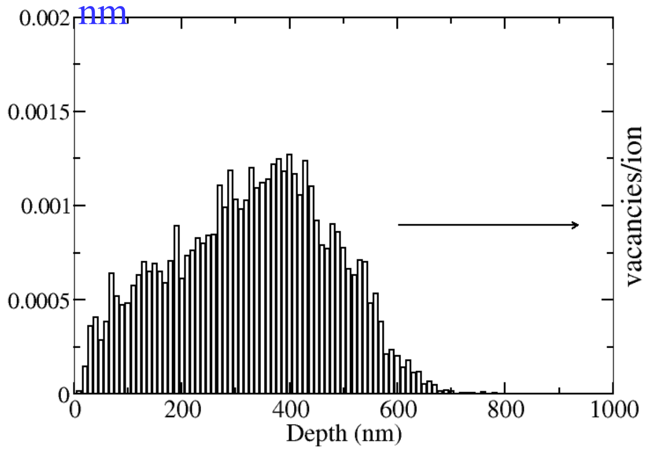
Loop diameter: **35 nm – 80 nm**

Grain Boundary: Less migration of defects to GB in contrast to Au

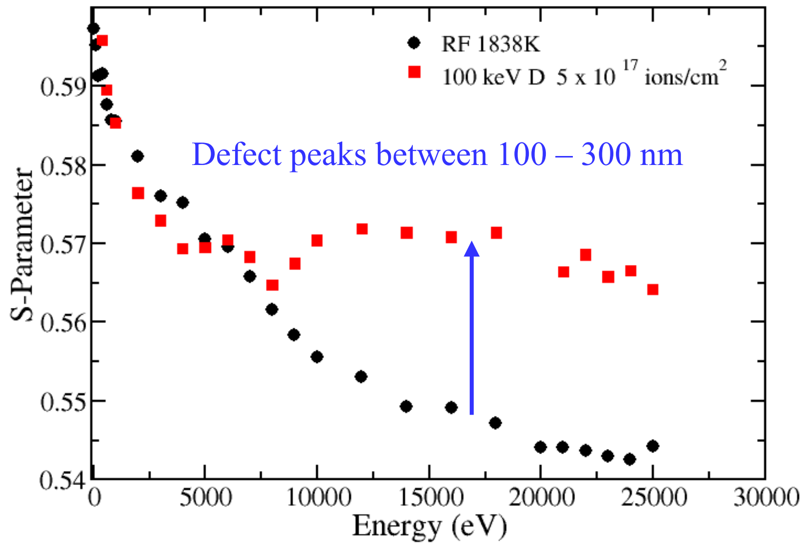
Low Energy (keV) Gaseous Ion Irradiation

Ions: 100 keV D^+; 250 keV He^+, Flux: 5.37×10^{14} $D^+/cm^2/s$ and 9.76×10^{12} $He^+/cm^2/s$;
Fluence 5×10^{17} D^+/cm^2 and 5×10^{15} He^+/cm^2 ; dpa = 0.005 and 0.03 for Ed = 90 eV

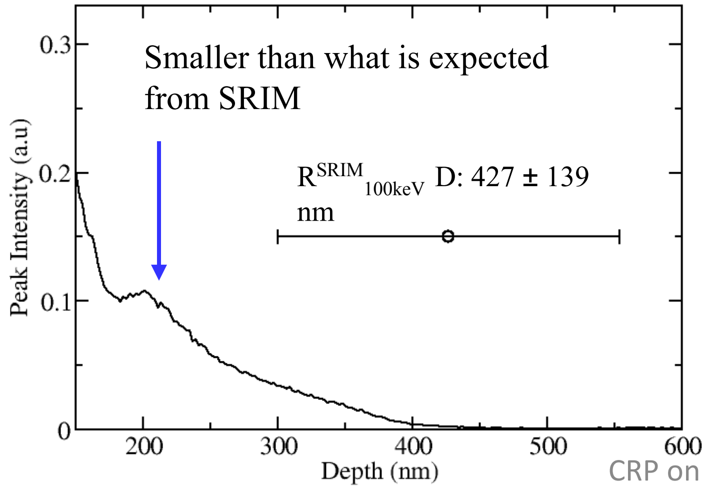
R^{SRIM}_{100keV} D: 427 ± 139 nm



Defect depth profile using positron DB spectroscopy

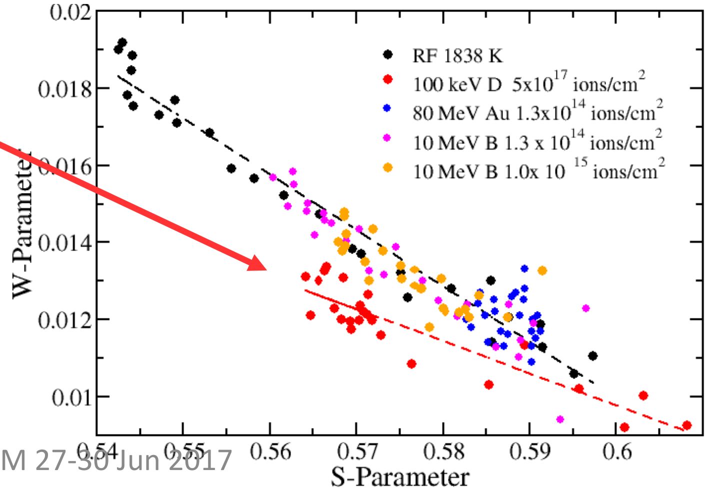


100 keV D depth profile - SIMS

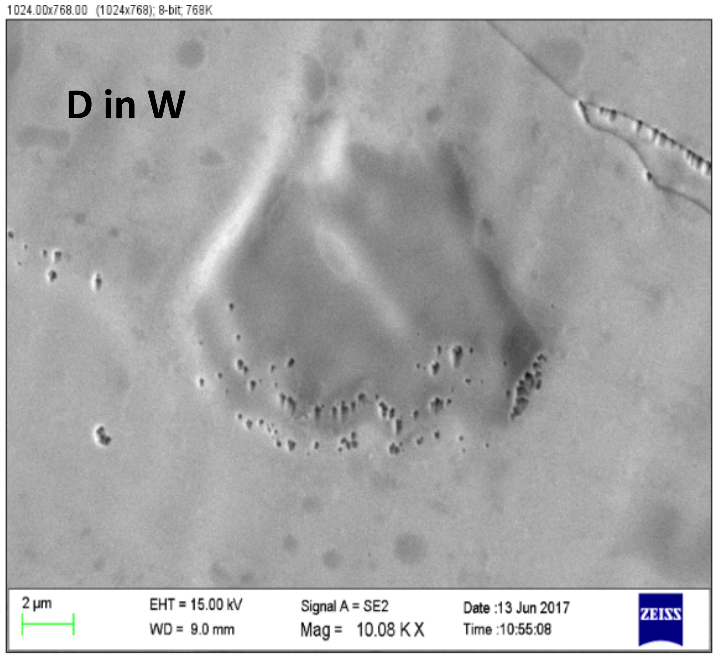


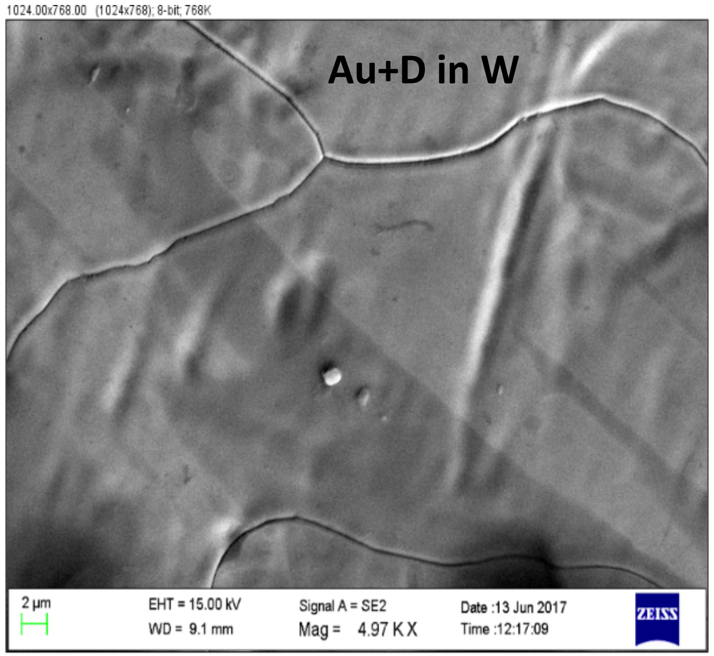
Annihilation from different core-electrons

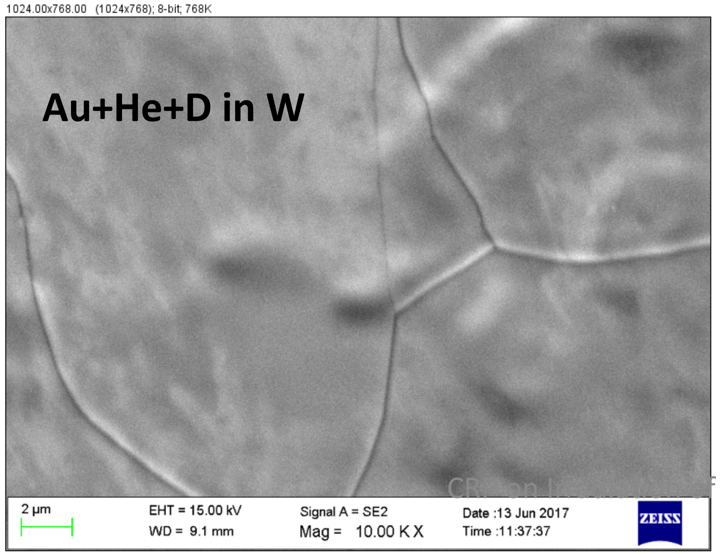
D-rich defects?



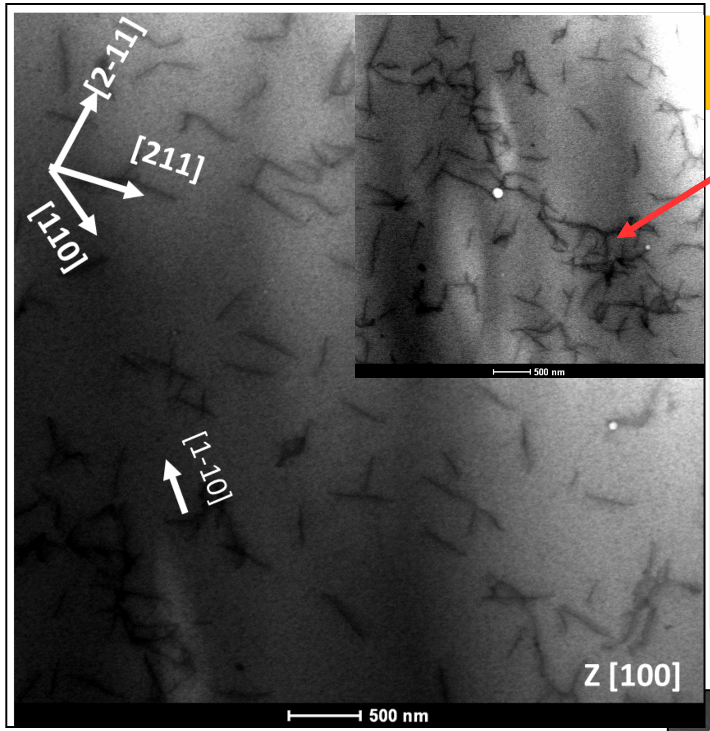
Blistering in D and He irradiated Tungsten







Blisters are seen in Deuterium irradiated, Deuterium in Au Pre-irradiated and Deuterium in Au and He pre-irradiated Tungsten



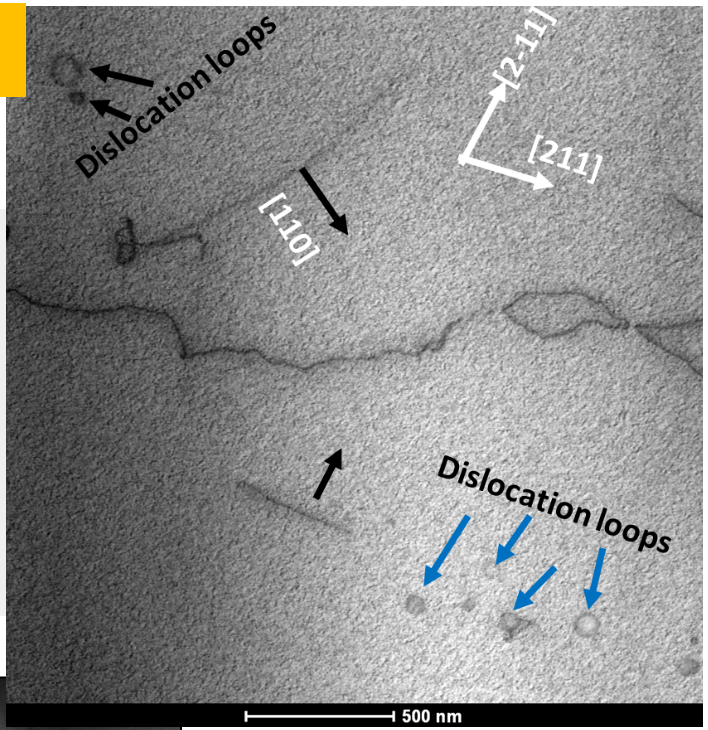
100 keV D

Dislocation network

Dislocation line length
= 120 nm – 1900 nm

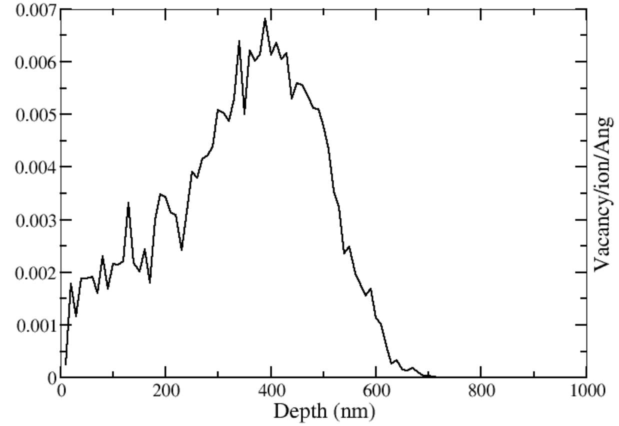
Dislocation loop dia
= 50 nm – 100 nm

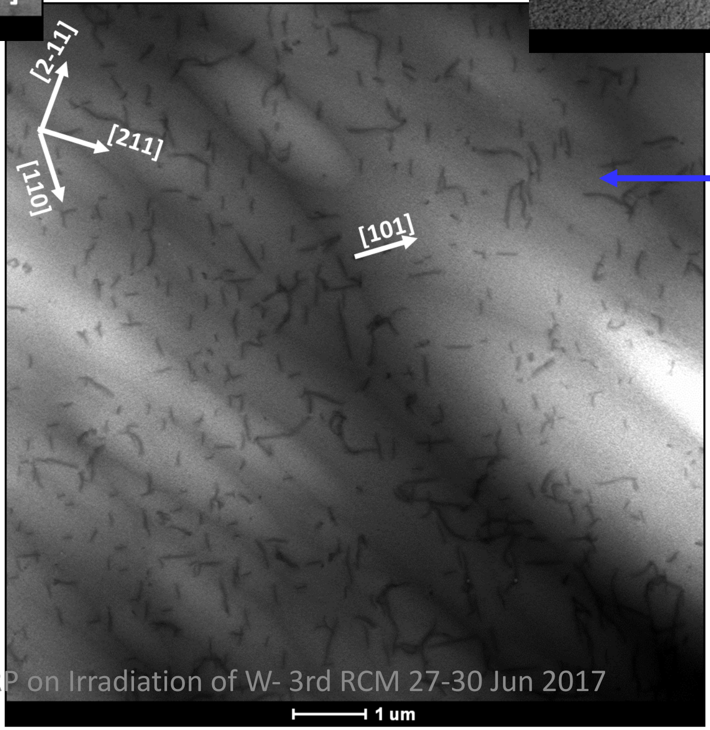
Dislocation density
= 9.7 x 10^{8} cm^{-2}



250 keV He

R^{SRIM}_{250keV} He: 429 ± 124 nm





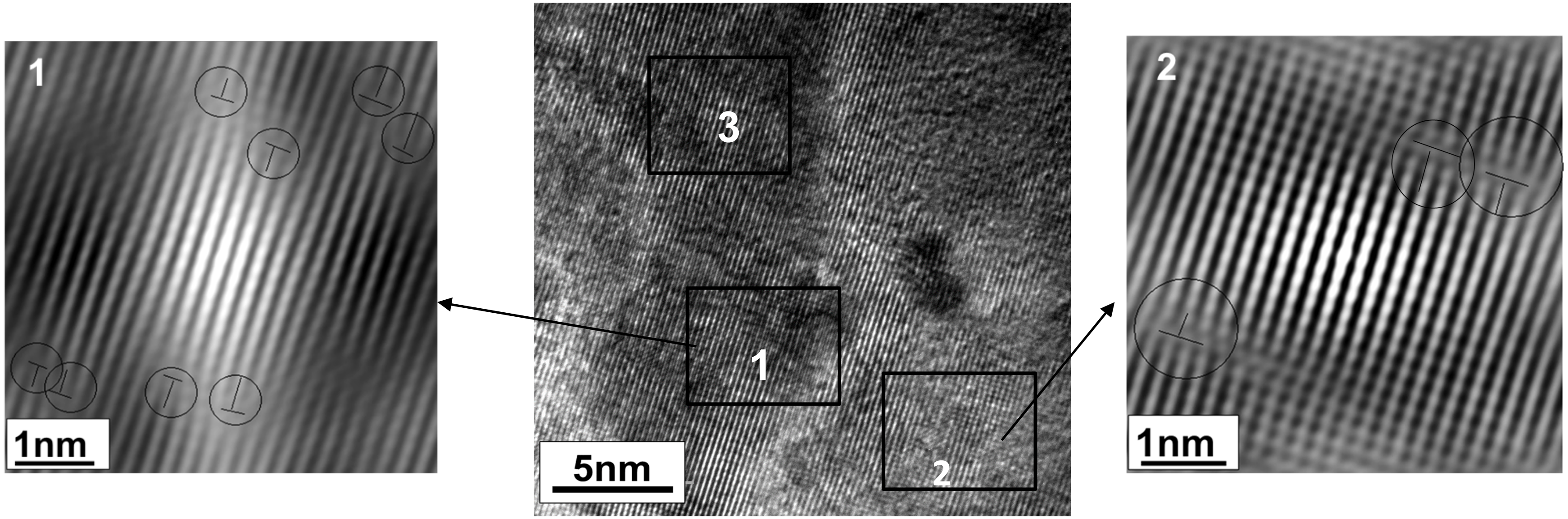
250 keV He

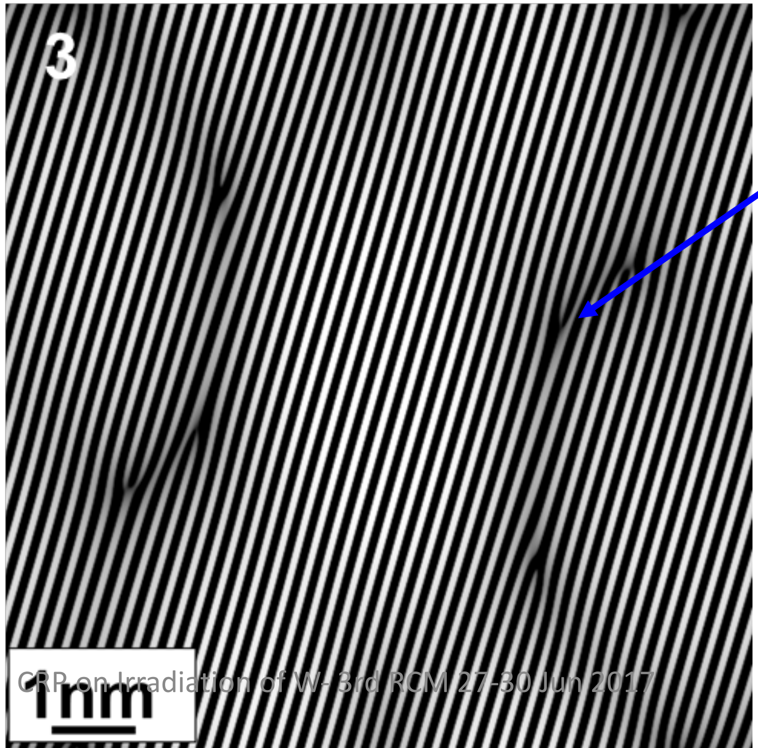
No loops were observed

Dislocation line length
= 40 nm – 1000 nm

Dislocation density
= 1.0 x 10^{9} cm^{-2}

HRTEM Images of He irradiated tungsten

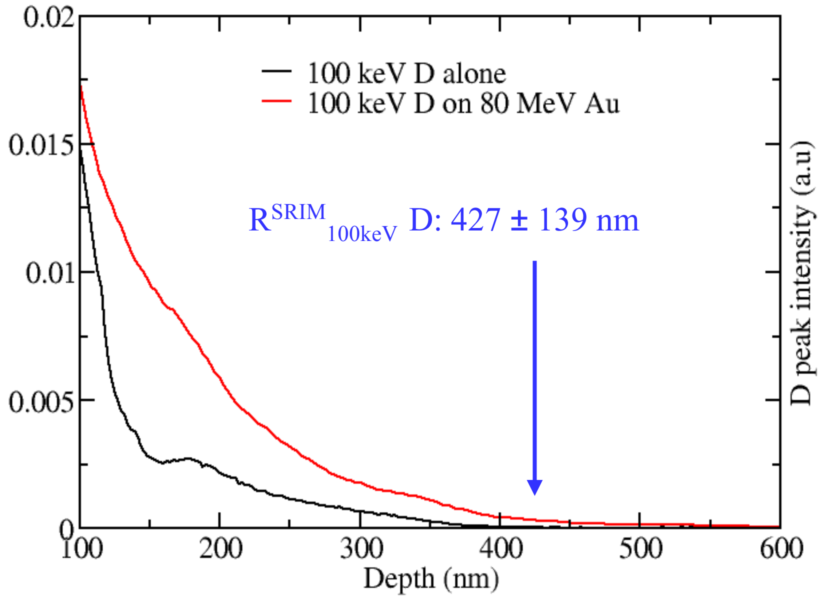




Edge dislocations

Inter-planar distance = 2.23 Å
→ (110) plane

Deuterium Depth Profile Measurements – SIMS – Preliminary Data

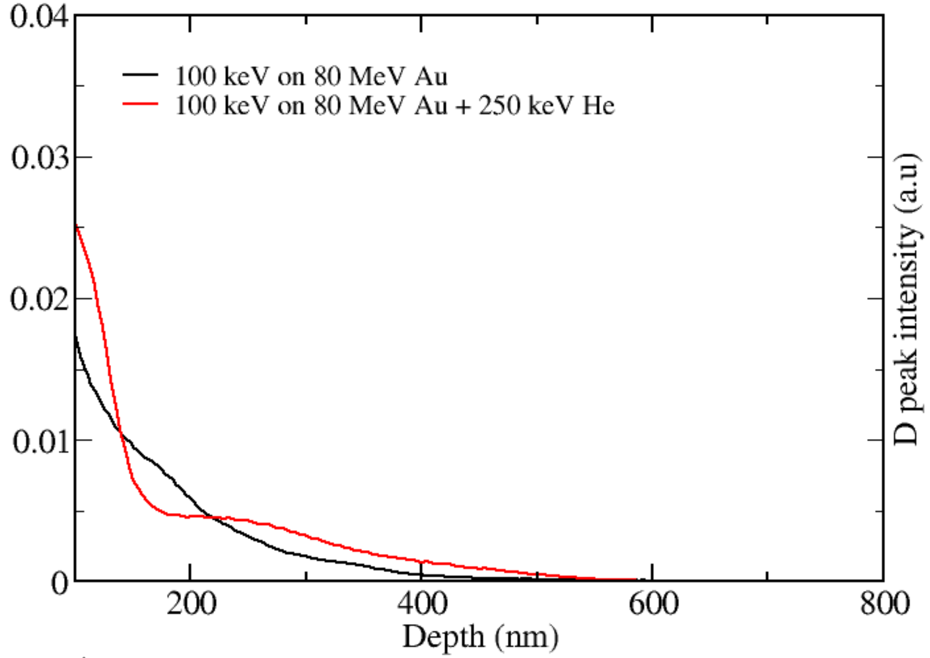


Helium seems to reduce the D trapping, even at this higher energies.

Ion driven permeation experiments showed that He reduces D permeation as well as trapping at 1-keV [H.T. Lee et.al, JNM 2011, 2014, Y. Hatano PSI 2016,V. Kh. Alimov 2012]

At higher energies, defect creation and trapping may be an additional factor as seen from TEM images

Surface roughness effects might be significant



Ongoing activities

Quantification of D-depth profiles using ERDA & SIMS

Quantification of He-depth profiles

Modelling of D depth profiles using trapping-detrapping models using MC simulations

Analysis of deuterium trapping in W-La_2O_3 alloys

Summary

- Modeling
  - MD simulations have been carried out to investigate the damage and cascade structure; observed splitting of cascades at high energy
  - While interstitials are located on the outer boundary of the cascade zone, vacancies seem to group near the core region
  - Cascade structure dependence on the choice of electronic-loss cut-off energy
  - Near-surface cascades show interstitials preferentially crowding at the top of the surface
- Experiments
  - Heavy & Light ion irradiation has been carried in recrystallized tungsten foils (80 MeV Gold, 10 MeV Boron, 250 keV He, 100 keV D)
  - Dislocations were seen in Au and B irradiated tungsten but the dislocation density is more for B as compared to Au
  - For D-irradiation, dislocation networks and loops were observed. Blisters were observed in cases of “only D”, “Au+D” and “Au+He+D”
  - PAS analysis shows that different types of defects including dislocations, point defects, vacancy clusters are present in heavy and light ion irradiated cases
  - He irradiation seems to locally deplete the D population in pre-damaged+D cases

Thank You